

How do the indoor size distributions of airborne submicron and ultrafine particles in the absence of significant indoor sources depend on outdoor distributions?

Abstract Although almost all epidemiological studies of smaller airborne particles only consider outdoor concentrations, people in Central Europe actually spend most of their time indoors. Yet indoor pollutants such as organic gases, allergens and dust are known to play a prominent role, often affecting human health more than outdoor ones. The aim of this study was to ascertain how the indoor particle size distributions of submicron and ultrafine particles correlate with the outdoor concentrations in the absence of significant indoor sources. A typical indoor particle size distribution pattern has one or two modes. In the absence of significant indoor activities such as smoking, cooking etc., outdoor particles were found to be a very important source of indoor particles. The study shows that in the absence of significant indoor sources, the number of indoor concentrations of particles in this size range are clearly lower than the outdoor concentrations. This difference is greater, the higher the number of outdoor concentrations. However, the drop in concentration is not uniform, with the decrease in concentration of smaller particles exceeding that of larger ones. By contrast, the findings with larger particle sizes (diameter $> 1 \mu\text{m}$) exhibit rather linear concentration decreases. The non-uniform drop in the number of concentrations from outdoors to indoors in our measurements considering smaller particles ($> 0.01 \mu\text{m}$) is accompanied by a shift of the concentration maxima to larger particle diameters.

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Practical Implications

The size distribution of submicron and ultrafine particles from outdoor air is different indoor. Consequently, epidemiological studies assessing the health impact of such particles must take into account these differences in particle size distribution.

Introduction

Numerous studies have reported links between airborne particles and a whole range of respiratory symptoms and pulmonary function deficits, emergency room visits and hospital admissions, cardiovascular symptoms and increased mortality (Abbey et al., 1995; Thurston et al., 1994; Wilson and Spengler, 1996). Because airborne particles do not constitute a uniform population, various types of particulate air pollution have been addressed in epidemiological studies (Phalen, 1998; Pope et al., 1995), the main ones being TSP (total suspended matter) and PM10s (particulate matter with an effective aerodynamic diameter less than $10 \mu\text{m}$). In recent years, many studies have

targeted fine particulate matter, i.e. particles with an aerodynamic diameter less than $2.5 \mu\text{m}$ (PM2.5s). PM2.5 is now generally regarded as the state of the art in the field of epidemiological studies owing to a number of indications that fine particles are more significant for human health than PM10s and larger particles (Murphy et al., 1998; Pope et al., 1995; Schwartz and Naes, 2000; Schwartz et al., 1996). Nevertheless, the ill-health effects of PM2.5 are still under discussion (Gamble, 1998; Murphy et al., 1998; Pekkanen et al., 1997; Tiittanen et al., 1999).

Only very few studies have paid special attention to the ill-health effects of smaller particles such as submicron and ultrafine particles (smaller than $1 \mu\text{m}$ and 100 nm , respectively). Moreover, the majority of

them are not concerned with epidemiological investigations, but linked to *in vivo* and *in vitro* experiments (Anderson et al., 1990; Churg et al., 1998; Li et al., 1999; Lundborg et al., 1999; Murphy et al., 1998; Osier and Oberdörster, 1997; Stone et al., 2000 etc). Some findings suggest that particle fractions significantly smaller than PM_{2.5} may be of great importance for human health (Burnett et al., 1995; Dreher et al., 1996; Oberdörster et al., 1995; Pekkanen et al., 1997; Peters et al., 1997; Tsai et al., 2000). Such particles may be particularly significant for airway diseases, including acute respiratory effects and cardiovascular effects among those exposed to them.

Recently, several measuring campaigns in the field of aerosol research have been carried out with respect to submicron and ultrafine particles, especially in urban areas (Franck and Herbarth, 2002; Hitchins et al., 2000; Jamriska et al., 1999; Morawska et al., 1999a,b etc). By contrast, an important knowledge gap persists with respect to indoor environments, including submicron and ultrafine particles. In many countries, the majority of people spend most of their time indoors, the figure for inhabitants of Central Europe ranging from 80% to more than 90%. Therefore, indoor particulates must be taken into account in environmental studies concerning the health impact of particulate matter. Although some studies have examined the health effects of indoor fine particulate matter (PM_{2.5}) (Jamriska et al., 1999; Leaderer et al., 1999; Lee and Chang, 1999; Neas et al., 1993), there is an almost complete dearth of information on the effects of indoor submicron and ultrafine particulates.

The primary importance of the indoor environment is generally accepted for airborne pollutants such as volatile organic compounds, polyaromatic hydrocarbons, other organic compounds, and many allergens. These indoor pollutants often affect human health more than outdoor ones. Furthermore, it has been established that outdoor and indoor particulates differ in terms of toxicity and mutagenicity (PM_{2.5}) (van Houdt et al., 1984; Monn and Becker, 1999).

Epidemiological studies in the field of submicron and ultrafine particles are usually forced to use outdoor particle concentrations because of the huge efforts connected with taking a large number of detailed size-resolved indoor particle measurements.

In view of the insufficient indoor data, the outdoor concentrations of submicron and ultrafine particles have to be regarded as the total burden accounted for by these particles. Of course, investigation must also be carried out into the superposition of indoor particle distributions by particulate generated by cooking, smoking and other sources.

The aim of this study was to ascertain how the indoor particle size distributions of submicron and ultrafine particles are correlated with the outdoor concentrations in the absence of significant indoor sources.

Methods

Measurements were carried out between summer 1999 and winter 2000/2001 using two differential mobility particle sizer systems simultaneously scanning the particle size distributions of particles between 15 and 800 nm. The systems were equipped with differential mobility analyzers (DMA) and condensation particle counters (CPC). The sample-to-sheath flow ratios amounted to 1:10 using sample flow rates of typically 0.2–0.3 l/min. A total of 1500 pairs of indoor and outdoor size distributions were used to compare indoor and outdoor situations. The indoor system was installed in a third-floor laboratory, and, for comparison, in a first-floor room (former gatehouse), both with the windows and doors kept closed and without active ventilation. The room volumes were approximately $5 \times 6 \times 3.5 \text{ m}^3$ and $5 \times 6 \times 3 \text{ m}^3$, respectively. There were no air-conditioning systems in either of the rooms.

To minimize the effect of indoor sources (Chao et al., 1998) during the experiments, the level of human activity was kept low.

The air change rate was estimated judging by the type of windows and the literature to be in the order of 0.2–0.4 per hour (Chao and Tung, 2001; Ihle, 1997; Koponen et al., 2001; Krooß et al., 1997; Reinmuth, 1996). The theoretical rate was calculated using the length of the joints of windows and doors to be 0.31 per hour and 0.33 per hour, respectively (DIN 4701). Additional measurements using SF₆ as tracer gas yielded a mean rate of 0.16 per hour (for method cf. Lohmeyer, 1992). According to the literature, the mean air change rate measured in apartments is 0.36 per hour. The lowest values were around 0.1 per hour, although with extremely airtight windows the rate may drop to just 0.02 per hour (Krooß et al., 1997).

The outdoor system was set up within the same campus on the roof of a building.

The campus is located in an urban area with no significant industry in the vicinity, and near a street with through traffic.

For analysis the data were exported via a spreadsheet calculation program to a statistical program system (STATISTICA 5.1 by Statsoft, Tulsa, OK, USA). All Pearson correlation coefficients > 0.1 and $< (-0.1)$ given in Figures 8–10 have P -values < 0.05 .

In the graphs (Figures 1, 2, 5–11), the particle diameters are plotted on a logarithmic scale.

Results and discussion

Concentration decrease

Figures 1 and 2 show examples of different shapes of indoor and outdoor size particulate distribution. The total indoor particle numbers are all lower than the

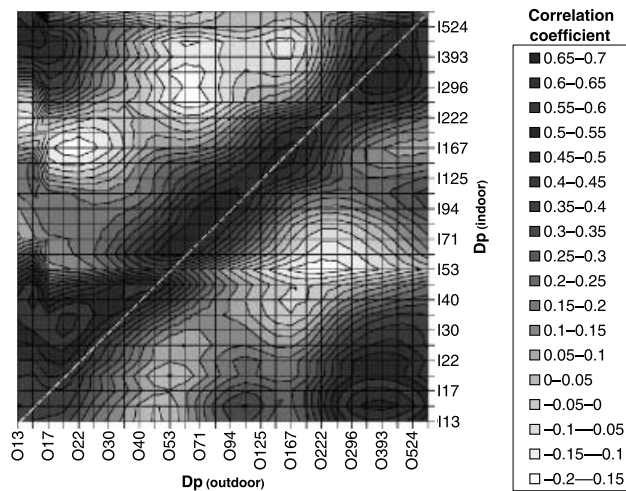


Fig. 1 Indoor and outdoor size distributions of particles (examples of paired outdoor and indoor measurements)

outdoor ones; between 6 and 78% of the total number of outdoor particles were found indoors. The highest ratios between indoor and outdoor concentrations were found at night in the absence of any human activity in the laboratory. These high ratios tend to correlate with low rather than high total outdoor particle numbers (Figure 3), which is also indicated by a negative Pearson correlation coefficient between the outdoor : indoor number concentrations and the outdoor total number concentration: -0.69 (P -value: < 0.0001). Most abundant in our measurements were indoor to outdoor total particle number of concentrations of 0.24 (Fig. 4). However, the concentrations of particles with different diameters vary (Figure 5), the strongest decrease being noted among Aitken-mode particles.

The concentration of particles with diameters exceeding $0.5 \mu\text{m}$ also decreases. This drop may be accounted for by the greater resistance of these particles' penetration of the indoor site. Compared

with smaller and larger sizes, particles approximately between 0.1 and $0.4 \mu\text{m}$ exhibit higher ratios of indoor to outdoor concentrations. In a few cases, in this diameter range the ratios exceed 1 (for examples, cf. Figure 6). A similar picture is given by the absolute differences of the outdoor and indoor particle numbers: Figure 7 shows smaller decreases for larger particle diameters.

Size changes

Looking at Figures 1 and 2, it can be seen that not only are the indoor concentrations lower than outdoors, but also that the concentration maximum has shifted to higher concentrations. This behavior is clearly evident in the mean particle number of concentrations throughout all measurements (Figure 8). Furthermore, whereas in the case of bimodal distributions the maximum of the smaller particle mode shifts to larger sizes, the maximum of the larger mode remains at nearly the outdoor diameter (Figure 1). Principally, this process can be evoked by capturing ultrafine particles in the indoor site and/or by the expansion of particles as they are transferred from the Aitken to the accumulation mode. Because of the occurrence of indoor to outdoor concentration ratios of a little more than 1 (see Figure 6) in the corresponding mode and the simultaneous decrease in the total number of particles, a transfer process cannot be excluded to contribute to the decrease in the number of Aitken particles. On the other hand, coagulation should not contribute significantly at these number of concentrations and residence times and measuring errors/weak indoor sources may well cause these indoor to outdoor concentration ratios of little more than 1.

A simple (constant) linear ratio between indoor and outdoor mass concentrations as measured for PM_{10} s and $\text{PM}_{2.5}$ s (Giess et al., 1998; Tsai et al., 2000; Wallace, 1996) was not observed for the

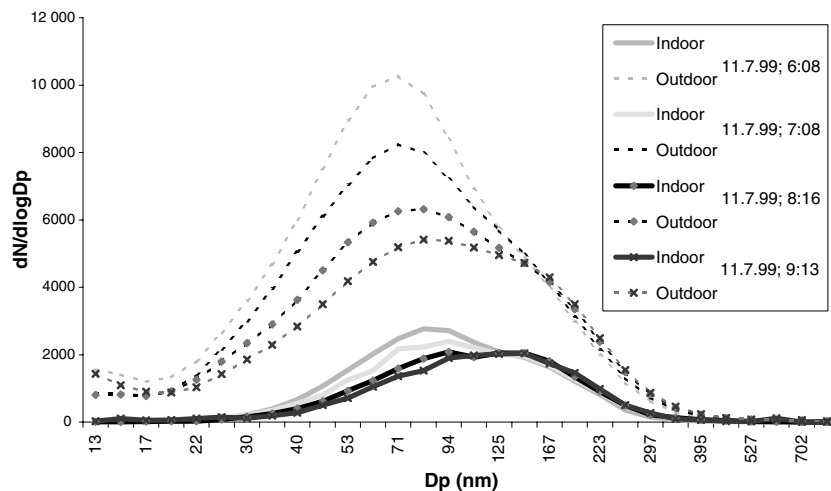


Fig. 2 Indoor and outdoor size distributions of particles (examples of paired outdoor and indoor measurements)

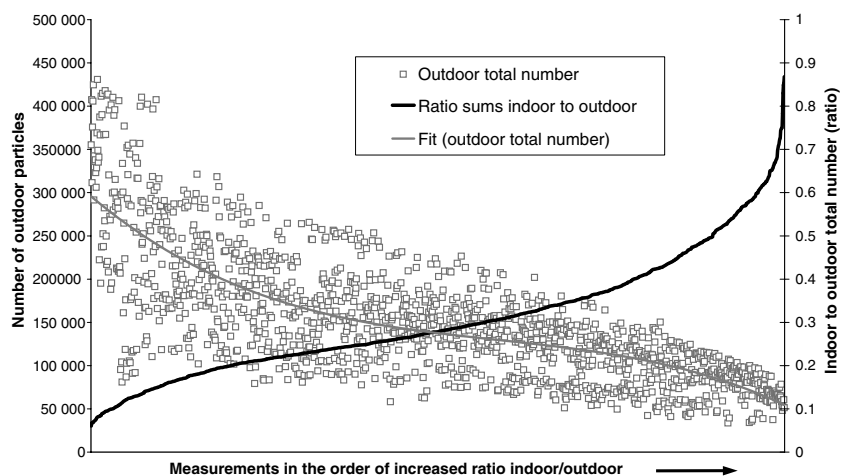


Fig. 3 Ratio of (indoor total particle number concentration):(outdoor total particle number concentration) [black line] vs. outdoor total particle number concentration [grey open squares]; fit: polynom of 3rd degree – grey line)

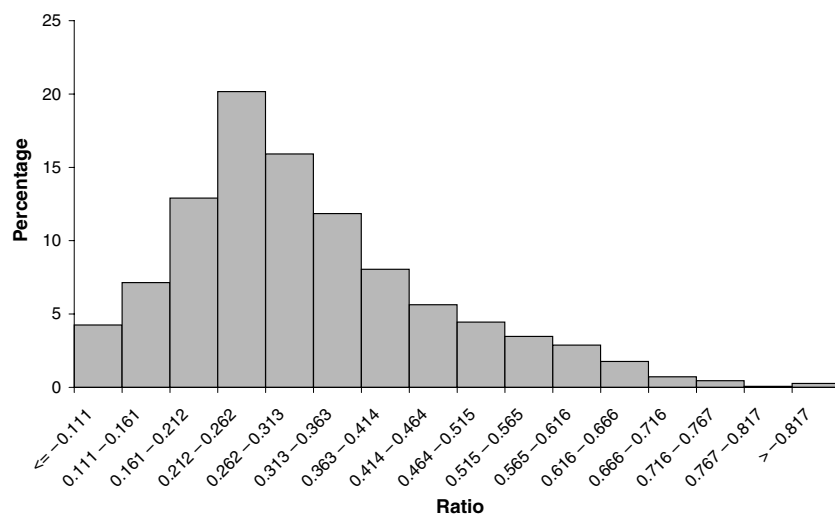


Fig. 4 Relative abundance of ratios of indoor to outdoor total number of concentrations within all measurements

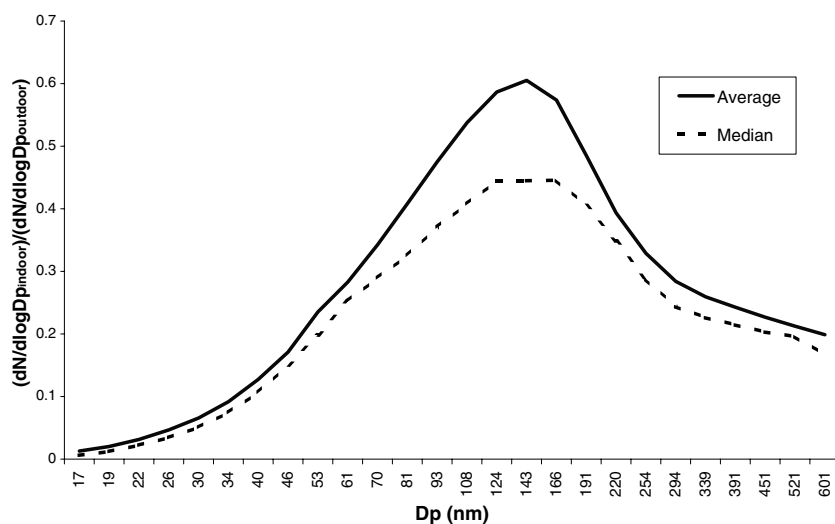


Fig. 5 Ratio of indoor/outdoor particle number of concentrations (average of all measurements in summer and winter time)

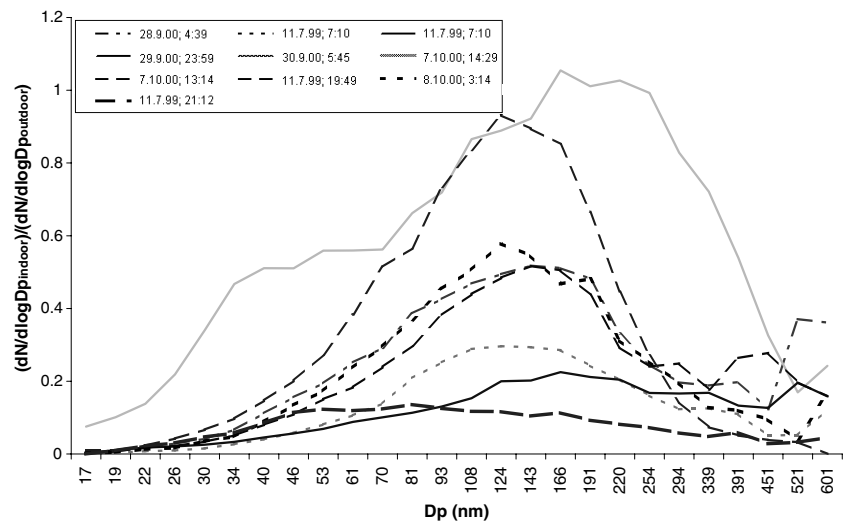


Fig. 6 Ratio of indoor/outdoor particle number of concentrations (examples of measurements)

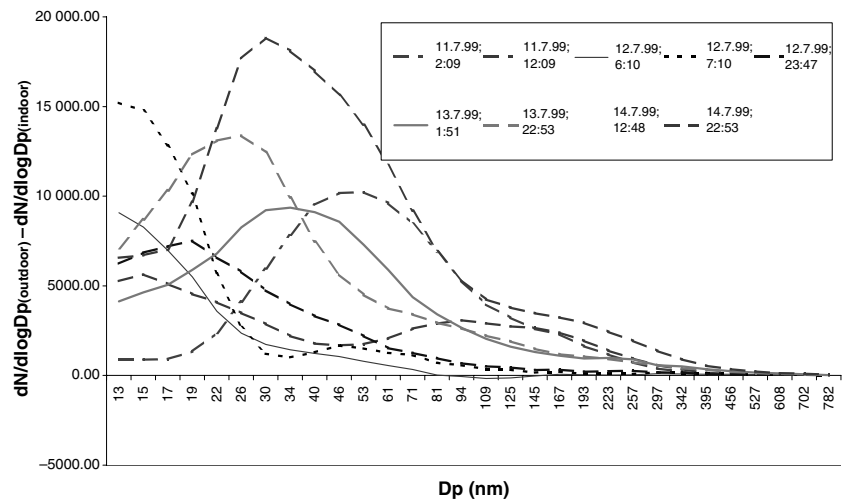


Fig. 7 Differences of outdoor minus indoor particle number of concentrations (examples of measurements)

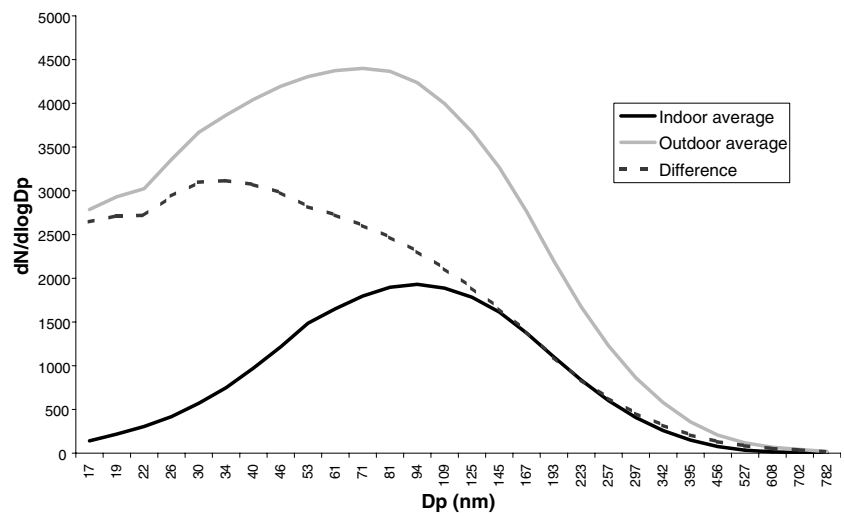


Fig. 8 Outdoor and indoor particle number of concentrations (average of all measurements in summer and winter)

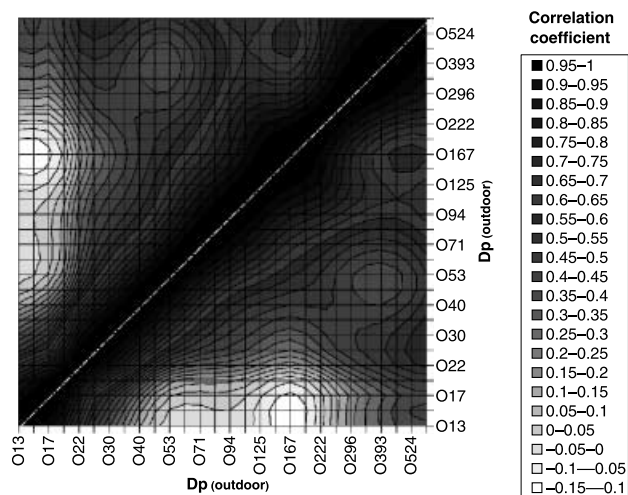


Fig. 9 Pearson correlation coefficients of outdoor number of concentrations of particles of given sizes with outdoor number of concentrations of particles of other sizes (bivariate histogram: x-axis: outdoor particle diameters; y-axis: outdoor particle diameters; z-axis: correlation coefficients, dark areas – high positive correlation)

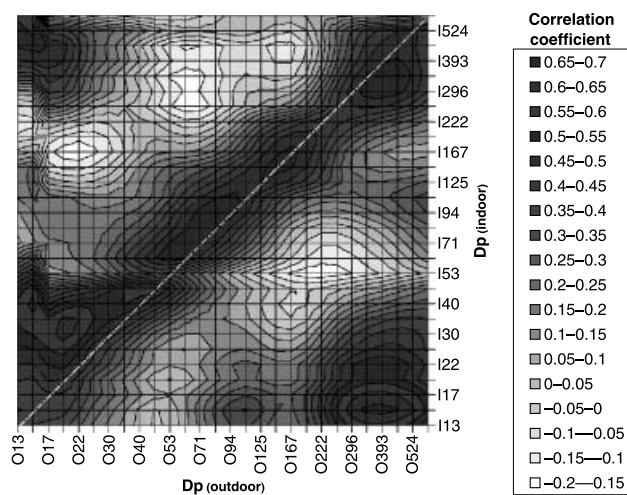


Fig. 11 Pearson correlation coefficients of indoor number of concentrations of particles of given sizes with outdoor number of concentrations of particles of other sizes (bivariate histogram: x-axis: outdoor particle diameters; y-axis: indoor particle diameters; z-axis: correlation coefficients, dark areas – high positive correlation)

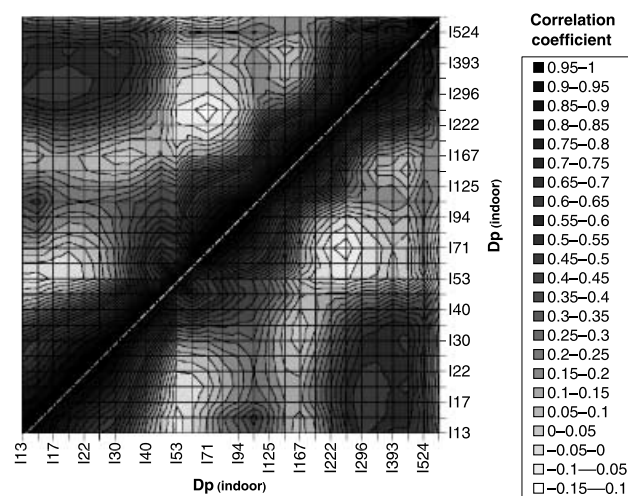


Fig. 10 Pearson correlation coefficients of indoor number of concentrations of particles of given sizes with indoor number of concentrations of particles of other sizes (bivariate histogram: x-axis: indoor particle diameters; y-axis: indoor particle diameters; z-axis: correlation coefficients, dark areas – high positive correlation)

particle number of concentrations of submicron and ultrafine particles.

Indoor–outdoor correlation

A nearly linear relationship was found by Jamriska et al. (1999) between the mass concentrations of PM_{2.5} and particles smaller than 0.7 μm in the time-dependence curves and by Giess (1998) for the indoor to outdoor particulate mass ratio of PM₇s. He found indoor/outdoor ratios of approximately 0.5–0.6.

Similar ratios have been stated for PM_{2.5}s (Jannseen et al., 1998; Li, 1994; Tsai et al., 2000; Wallace, 1996) estimated the relation between personal, indoor, and outdoor air concentrations of PM₁₀s, and ascertained that personal particle concentrations exceed the others. Monn et al. (1997) also found an indoor to outdoor ratio of 0.7 for PM₁₀s.

To see the dependence of the number of concentration of indoor particles of a selected diameter on the concentrations of the indoor particles of other diameters as well as on the concentrations of the outdoor ones, we calculated the correlation coefficients between the number of concentrations of particles of different diameters.

The diameters of both indoor and outdoor particles are best correlated with diameters of other indoor and outdoor particles which differ only slightly (Figures 9 and 10). Negative correlation reflects the decrease in the frequency of distinct particle sizes if other sizes are frequent. The patterns of correlation coefficients between indoors and outdoors differ most markedly with respect to these negative coefficients.

The correlation pattern between the number of outdoor and indoor particle concentrations at a given diameter indicates that the number of indoor concentrations of given sizes are correlated with the outdoor such that in the case of outdoor particles with diameters less than approximately 250 nm, the indoor particle concentrations of given sizes correspond to concentrations of smaller outdoor particles (Figure 11).

The presence of indoor sources such as smoking or cooking means that indoor size distribution patterns are mainly determined by these sources. In this case, the correlation patterns mentioned do not exist.

Conclusions

Because humans spend 80% or even more than 90% of their time indoors, the resulting burden arises from a suitably weighted average emphasizing indoor particulates. Outdoor particles deliver a remarkable contribution to indoor concentrations. Therefore, in default of actual indoor measurements, outdoor particle size distributions can be used in epidemiological investigations to substitute for actual particle size distributions.

The indoor environment is generally shielded against outdoor particulates, leading to lower number of concentrations, indoors than outdoors. Unlike the decrease in mass concentrations of larger particle fractions (Giess, 1998; Jamriska et al., 1999; Jannseen et al., 1998; Li, 1994; Monn et al., 1997; Tsai et al., 2000; Wallace, 1996), the decrease in the number of concentrations of submicron and even more ultrafine particles is not linear. Therefore, to assess the health impact of these particles, the different decrease in the numbers of different particle sizes must be taken into

account if outdoor concentrations are used instead of indoor ones. Furthermore, the number of concentration maxima of indoor particles is shifted to larger diameters with respect to outdoor particle concentration. The higher the total number of outdoor concentration, the lower the total number of concentration of indoor particles.

On the other hand, the importance of indoor sources can be seen by comparing measurements by the authors in different habitations using the same methods, and has also been described by other authors employing different methods (van Houdt et al., 1984; Leaderer et al., 1999; Lee and Chang, 1999; Li, 1994; Monn et al., 1997; Tsai et al., 2000 etc). Consequently, if indoor data are unavailable, significant indoor sources should be avoided, or special attention should at least be paid to the possible effects of such sources. A more extensive study focusing on meteorological conditions, seasonal variations, and small-scale spatial variations has been started in an attempt to further clarify this topic. It needs to be augmented by other studies considering different types of indoor sources.

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