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Abstract

The number of ultrafine particles in urban air may be more health relevant than the usually measured mass of particles smaller than 2.5 or 10 μm . Epidemiological studies typically assess exposure by measurements at a central site. Limited information is available about how well measurements at a central site reflect exposure to ultrafine particles.

The goals of this paper are to assess the relationships between particle number (PN) and mass concentrations measured outdoors at a central site, right outside and inside the study homes. The study was conducted in four European cities: Amsterdam, Athens, Birmingham and Helsinki. Particle mass (PM_{10} and $\text{PM}_{2.5}$), PN, soot and sulfate concentrations were measured at these sites. Measurements of indoors and outdoors near the home were made during 1 week in 152, mostly non-smoking, homes. In each city continuous measurements were also performed at a central site during the entire study period.

The correlation between 24-h average central site outdoor and indoor concentrations was lower for PN (correlation among cities ranged from 0.18 to 0.45) than for $\text{PM}_{2.5}$ (0.40–0.80), soot (0.64–0.92) and sulfate (0.91–0.99). In Athens, the indoor–central site correlation was similar for PN and $\text{PM}_{2.5}$. Infiltration factors for PN and $\text{PM}_{2.5}$ were lower than for sulfate and soot. Night-time hourly average PN concentrations showed higher correlations between indoor and central site, implying that indoor sources explained part of the low correlation found for 24-h average concentrations.

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Measurements at a central site may characterize indoor exposure to ambient particles less well for ultrafine particles than for fine particle mass, soot and sulfate.

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1. Introduction

Numerous epidemiological studies have documented that current day levels of particulate matter (PM) air pollution are associated with adverse health effects including respiratory symptoms, lung function, hospital admissions for cardio-respiratory disease and mortality (Brunekreef and Holgate, 2002; US Environmental Protection Agency, USEPA, 2004). In most recent epidemiological studies PM has been characterized as the mass concentration of particles with an aerodynamic diameter less than 10 µm (PM₁₀) or less than 2.5 µm (PM_{2.5}). Therefore air quality standards or guidelines are currently expressed as PM₁₀ and/or PM_{2.5}. Ambient PM is a mixture of particles that may differ widely in chemical composition and size (Brunekreef and Holgate, 2002; USEPA, 2004; Sioutas et al., 2005). Coarse particles (>2.5 µm) are mainly formed by mechanical processes and fine (<2.5 µm) and ultrafine particles (UFP) (<0.1 µm) are directly emitted (e.g. diesel soot) and formed by chemical reactions from gaseous precursors. Although the health effects associated with PM exposure are generally known, the mechanisms and the fractions of PM that are responsible for the observed effects are less known. Soluble transition metals, strong aerosol acidity and UFP have been proposed as components responsible for observed health effects (Brunekreef and Holgate, 2002). Seaton et al. (1995) have hypothesized that the *number of UFP* is a more relevant exposure variable than the *mass* of PM because UFP have a large surface area and can penetrate into the interstitium and into the blood stream. A better characterization of the health relevant particle fraction will have major implications for air-quality policy since it will determine which sources should be controlled.

Epidemiological studies of acute effects of air pollution have typically used the outdoor concentration measured at one or a few fixed sites to characterize daily exposure to PM. A critical assumption is that the variation in time of the

concentration measured at the fixed site reflects the variation in time in the personal exposure of people living in the area. A poor correlation between outdoor and personal exposure could result in seriously biased estimates of the health effect of a pollutant from epidemiological studies. Several studies have documented that this time series correlation is generally high for PM₁₀ and especially PM_{2.5} (Brunekreef et al., 2005 and references therein). For UFP, no information is available about the relationship between central site concentrations and personal exposure (Pekkanen and Kulmala, 2004). The spatial heterogeneity of outdoor concentrations and the indoor/outdoor relationships may be different for UFP number concentration compared to PM_{2.5} mass. Sioutas et al. (2005) have identified the need to collect more information on infiltration of (ultrafine) particles in relevant indoor environment as an important research topic. Information about indoor/outdoor relationships for UFP is available from studies in a few offices or homes (Koponen et al., 2001; Cyrus et al., 2004; Patterson and Eatough, 2000; Abt et al., 2000a; Long et al., 2000). These studies were typically conducted under unoccupied conditions. Outdoor concentrations were usually measured directly outside the home, whereas in most epidemiological time series studies measurements are conducted at a central site. These works have provided useful insights into mechanisms affecting the relationship between indoor and outdoor particle concentrations. However, the observed correlations and infiltration factors cannot readily be used to assess population exposures to UFP.

The current paper addresses the relationships between indoor and outdoor concentrations of particle number (PN), particle mass and composition (soot and sulfate) in four major European cities. The goals of the paper are to assess (a) the correlation between indoor and (central site) outdoor concentrations and (b) the indoor/outdoor regression slopes for PN, mass and specific components. Improvement of exposure assessment for UFP in epidemiological studies was an important

overall objective of the study. Analyses of the spatial variation have been reported elsewhere (Puustinen et al., 2007).

2. Material and methods

2.1. Study design

The study was performed within the framework of the multi-center Relationship between Ultrafine and fine PM in Indoor and Outdoor air and respiratory Health (RUIOH) study. The study was conducted in four European cities, namely, Amsterdam (The Netherlands), Athens (Greece), Birmingham (United Kingdom) and Helsinki (Finland). These four cities were selected to cover different areas of Europe and because of expertise with PN monitoring.

Particle mass (PM_{10} and $PM_{2.5}$) and number concentrations (PN), soot and sulfate (SO_4^{2-}) concentrations were measured. A central site was selected in each city, where continuous measurements were performed during the entire RUIOH campaign. Measurements outdoors (directly outside the home) and inside the homes of 35 patients with asthma or Chronic Obstructive Pulmonary Disease (COPD) were made in each city to evaluate spatial variability and indoor/outdoor relationships. In Amsterdam, 15 additional homes located on the main road network were selected with additional local funding. Only homes with few indoor sources were selected, as the main goal was to assess the extent to which particles from outdoor origin affect indoor air quality. Indoor sources may confound the ratio between indoor and outdoor particles and their correlation. In each home, measurements were conducted during 1 week. Because of limited availability of equipment, measurements were taken one home at a time. The study period was from October 2002 to March 2004. Standard questionnaires were used to characterize the indoor and outdoor sampling locations with respect to distance to the city center, traffic intensity, home characteristics and presence and use of indoor sources. In all cities the same type of samplers and the same standard operating procedures (SOP) for air monitoring were used. In all cities, medical ethical approval was obtained for the study from the respective medical ethical committees. All participants signed informed consent forms.

2.2. Study areas and study homes

Study areas are described in more detail elsewhere (Puustinen et al., 2007). Briefly, in each city a central site was selected that was an urban background location located reasonably centrally in the study area. Study homes were chosen from the entire metropolitan area, as this is common practice in epidemiological time series studies. Subjects had to be older than 35 years of age, diagnosed with asthma/COPD and preferably non-working or work less than 16 h per week outside their house.

To make sure the indoor/outdoor measurements were representative for the study area, different home types were selected (e.g. single-family homes, apartments). Homes with few indoor sources were selected. Non-smoking households had to be selected, that is both the study subjects and his/her family should not be regular smokers. No further restrictions were applied. In Amsterdam and Birmingham, it was not possible to exclude homes with gas cooking, which would have excluded the majority of homes.

Indoor measurements were conducted in the living room. Samples were collected at a height of 1–2 m, at least 2 m away from doors, windows, ventilation inlets and potential indoor sources (e.g. geyser, gas range); more than 20 cm away from a wall and never above a heating source. The sampling inlets of two samplers were placed more than 1 m apart and at about the same sampling height (± 50 cm). Sampling inlets were located at least 1.5 m away from any pump outlet and in a place that did not disturb the inhabitants. Residential outdoor measurements were conducted right outside the home, usually on a balcony or in the garden.

2.3. Monitoring methods

2.3.1. Particle number

Monitoring methods and procedures have been described in detail (Puustinen et al., 2007). Briefly, in all four cities continuous PN concentrations were measured with condensation particle counters (CPC model TSI 3022A, TSI, St. Paul, MN, USA). The CPC 3022A effectively counts the number concentration of particles between 7 nm and 3 μ m. Measurements were made according to a common SOP. The SOP included requirements for the sampling configuration (length and material of sampling lines), CPC settings (e.g. high flow rate), checking

of the proper functioning of the instruments, quality control and data management. To make indoor measurements feasible, the instrument was placed in a sound-insulation box and butanol odor was removed by connecting an active carbon cylinder to the outlet of the CPC followed by an absolute filter to remove carbon particles from the exhaust air. In Helsinki and Athens one CPC was used for monitoring inside and directly outside the home. A sample changer was used to switch sampling between indoor and outdoor air. In Amsterdam and Birmingham two CPCs were used to measure outdoor and indoor air of the study homes. We calculated hourly and 24-h average PN concentrations from the 1-min data, if at least 66% of the hour/24-h time period had valid data. Twenty-four hour averages were calculated from noon to noon, coinciding with the PM_{2.5} and PM₁₀ sampling period.

Monthly intercomparisons of the instruments used in a city were performed. Most CPC intercomparisons showed that the absolute concentrations of the different instruments agreed within 20%. There were a few intercomparisons in Greece and the UK with very poor agreement (no correlation and up to a factor of two differences in absolute levels). In all cases this was traceable to equipment problems in one or two of the CPCs, which were then taken to the laboratory for maintenance. Field data from these CPCs were deleted from the database.

2.4. PM₁₀, PM_{2.5} soot and sulfate

Twenty-four hour average concentrations of PM₁₀ and PM_{2.5} were measured with Harvard impactors operating at 101 min⁻¹ (Air Diagnostics and Engineering Inc., Naples, ME, USA). SOPs provided directions for sampling, weighing and reflectance measurements. These procedures have been described before (Janssen et al., 2000; Brunekreef et al., 2005). Briefly, particles were collected on 37 mm 2 µm pore size Teflon filters. Filters were changed generally between 11 and 13 h. To limit losses of semi-volatile components, we stored the filters immediately after sampling in a refrigerator at 4 °C until weighing. Losses during sampling and condensation of gases (Subramanian et al., 2004) on the (generally low loadings of) collected particles cannot be prevented with the standard methods used to measure PM_{2.5}. Particle mass was determined by gravimetric analysis using analytical

microbalances with a precision of 1 µg. Teflon filters were conditioned for 48 h in stable temperature (21 ± 0.5 °C) and relative humidity (35 ± 5%) conditions before pre- and post-weighing.

Reflectance of all PM_{2.5} filters was measured using smoke stain reflectometers. Reflectance was transformed into absorbance, following the ISO 9835:1993 standard. Various studies (Cyrus et al., 2004; Brunekreef et al., 2005 and references therein) have shown that absorbance is a good surrogate for elemental carbon (here referred to as soot).

The sulfate content of 50% of the central site and 50% of the indoor PM_{2.5} samples was determined with ion chromatography using procedures described before (Brunekreef et al., 2005). All samples were analyzed in one central laboratory using a Dionex DC-100 ion chromatograph (Sunnyvale, CA, USA), after extraction in 10 ml ultrapure water. Sulfate measurements were conducted after the fieldwork was completed. The analysis provided concentrations of sulfate, nitrate, nitrite, chloride and phosphate. Because of sampling losses of nitrates due to volatilization, we focus on the sulfate data. From each subject we targeted to select four non-consecutive indoor samples and the corresponding central site samples in order to maximize the information content.

Quality assurance/control included collection of field blanks, field duplicates and an intercomparison of the weighing and reflectance measurements of the four laboratories. Average field blanks were subtracted from all measurements. Detection limits (calculated as three times the standard deviation of field blanks) for PM_{2.5} and PM₁₀ ranged from 1.1 to 4.7 µg m⁻³. The highest value was obtained in Athens, where due to the high concentrations no measurements were below this detection limit. The coefficient of variation (CV) calculated from field duplicates ranged from 6% to 9% for PM_{2.5} and PM₁₀; 9–24% for coarse particles (PM₁₀–PM_{2.5}) and 5–10% for soot across the four cities.

2.5. Data analysis

The main calculations were performed for 24-h average concentrations, because this is the main exposure variable in epidemiological time series studies and comparison between PN and PM₁₀/PM_{2.5}—available as 24-h averages—was an important objective. For PN, the availability of hourly data allowed us to assess diurnal variability and night-time

hours (from 0:00 to 6:00), assuming minimal impact of indoor sources.

The correlation between indoor and outdoor measurements was first calculated for each home separately. Next the distribution of individual correlation coefficients was evaluated, with the main interest in the median. Both Spearman and Pearson correlation coefficients were calculated. Only homes for which three or more valid measurements were obtained were included.

Indoor air pollution concentrations are affected by infiltration of outdoor pollutants into the home and indoor sources. To quantify the impact of outdoor air and indoor sources on indoor air quality, various studies have used mass balance model approaches (Meng et al., 2005; Polidori et al., 2007; Hänninen et al., 2004). Assuming steady conditions and considering the home as one well-mixed compartment, the following equation can be used:

$$C_{\text{in}} = (P AER C_{\text{out}})/(AER + k) + Q_{\text{is}}/(AER + k)V \\ = F_{\text{inf}}C_{\text{out}} + C_{\text{ig}},$$

where C_{in} and C_{out} are the concentrations of particles indoors and outdoors respectively ($\mu\text{g m}^{-3}$); P is the penetration factor (dimensionless),

AER the air exchange rate (h^{-1}), k the decay rate (h^{-1}), Q_{is} the emission due to indoor sources ($\mu\text{g h}^{-1}$) and V the home volume (m^3); F_{inf} is the infiltration factor and C_{ig} the particle concentration generated indoors. The infiltration factor can be interpreted as the fraction of ambient particles that penetrate indoors and remain airborne. Based upon this formula we estimated the infiltration of outdoor particles by linear regression analysis: the slope can be interpreted as the infiltration factor and the intercept as indoor generated particles (Meng et al., 2005; Hänninen et al., 2004; Ho et al., 2004). Since we had repeated observations per home, we first calculated regression slopes and intercepts per home and then summarized the distribution with the median.

3. Results

3.1. Description

Measurements were conducted between October 2002 and March 2004 in 152 homes in the four cities (Table 1). The characteristics of the homes differed substantially across cities. In Amsterdam more homes were defined as a traffic site and more homes

Table 1
Description of homes included in indoor–outdoor monitoring

	Helsinki	Athens	Amsterdam	Birmingham
Number of homes	37	35	50	30
Distance homes to central site (km) ^a	7.6 (2.5–34)	6.2 (0.7–18.8)	3.3 (0.3–9.5)	7.0 (0.9–29.7)
City center sites (<i>n</i> , %)	6 (16%)	9 (26%)	31 (62%)	1 (3%)
Site type				
Urban background (<i>n</i> , %)	32 (87%)	22 (63%)	28 (56%)	23 (77%)
Traffic (<i>n</i> , %)	5 (13%)	13 (37%)	22 (44%)	7 (23%)
<i>Home characteristics</i>				
Apartment (<i>n</i> , %)	26 (70%)	33 (94%)	36 (72%)	3 (10%)
Age home (<i>n</i> , %)				
< 1945	4 (11%)	0 (0%)	29 (58%)	13 (43%)
1945–1980	25 (68%)	20 (57%)	12 (24%)	14 (47%)
1980–2000	7 (19%)	14 (40%)	9 (18%)	2 (7%)
2001–2002	1 (3%)	1 (3%)	0	1 (3%)
Home volume (mean, m ³)	202	254	305	357
<i>Indoor sources/ventilation</i>				
Smoker in the home (<i>n</i> , %)	0 (0%)	6 (17%)	2 (4%)	4 (13%)
Smoking reported (<i>n</i> , %) ^b	4 (0%)	139 (1%)	173 (1%)	118 (1%)
Gas for cooking (<i>n</i> , %)	1 (3%)	4 (11%)	47 (94%)	20 (67%)
Open kitchen (<i>n</i> , %)	10 (27%)	11 (31%)	15 (30%)	2 (7%)
Mechanical ventilation/air conditioning (<i>n</i> , %)	5 (14%)	13 (37%)	1 (2%)	1 (3%)

^aMedian with minimum–maximum in parentheses.

^bNumber (%) of half-hour periods with smoking by anyone in the living room reported.

were located in the city center than in the other cities. This was due to inclusion of the 15 homes along the main (inner city) road network of Amsterdam. The majority of homes were apartments, except in Birmingham. Consistently, the average home volume of the selected homes in Birmingham was the largest. Very few new homes were selected in Amsterdam and Birmingham. In both cities there was a substantial fraction of pre 1945 homes. Because of the selection procedures, smoking occurred only in few homes. In all cities, smoking occurred during no more than 1% of the measurement week. Gas was used for cooking in the majority of homes in Amsterdam and Birmingham, whereas it was very uncommon in Athens and Helsinki. Permanent mechanical ventilation or air conditioning in the living room was present infrequently, except in Athens.

Valid measurements for at least 1 day were obtained in most of the 152 homes. For $PM_{2.5}$ valid measurements were obtained in 147–151 homes (range for central site outdoor, residential-outdoor and home indoor). For PN valid measurements were obtained for 133 homes (directly outside the home) and 144–146 homes (indoor and central site). The relatively large number of missing data for the residential-outdoor measurements occurred mostly in Birmingham (12 homes), because of problems with the CPC. The average number of valid measurement days per home was close to the designed seven, with the exception of the central site PM_{10} and $PM_{2.5}$ measurements in Birmingham and Helsinki (average 5.9). There was a small number of homes with less than three measurements per home. Fewer sulfate measurements were available because 50% of the $PM_{2.5}$ samples were analyzed.

The current study involved measurements of indoor/outdoor relationships for PN concentrations in a much larger number of homes than in previously published studies. Most previous studies involved measurements in a single office (Koponen et al., 2001), single hospital (Cyrys et al., 2004), single school (Patterson and Eatough, 2000) or a few homes (Abt et al., 2000a, b; Long et al., 2000; Morawska et al., 2001) or mixture of homes/offices (Matson, 2005). Another difference is that the measurements were performed under normal living conditions, whereas previous studies typically performed measurements under controlled conditions. The advantage of controlled conditions is that the impact of specific factors (e.g. air exchange rate, a

specific indoor source) can be studied with less confounding by other factors. The disadvantage is that due to the small number of homes and the artificial conditions, the quantitative relationships cannot be readily used to assess population exposures relevant for epidemiological studies and risk assessment. Although the current study included a much larger set of homes and although we attempted to select a relevant variety of homes, we do not claim that the selection of homes was representative for the entire housing stock of the four cities. We did, however, succeed in constraining our measurements to homes without active smokers to avoid the swamping effects of environmental tobacco smoke.

Concentrations measured at central, residential and indoor locations differed substantially between cities (Fig. 1a–f). Indoor concentrations were generally lower than the outdoor concentrations. The city with the lowest PN concentration at the central site (Helsinki) were also characterized by the lowest indoor concentrations. The cities with the highest PN at the central site and directly outside the homes were Athens and Amsterdam, respectively. The indoor concentrations in Athens did not differ significantly from the Amsterdam indoor concentrations. For $PM_{2.5}$ the pattern across cities in mean central site concentrations was also reflected in mean indoor concentrations. Especially indoors, a few very high concentrations of PN, $PM_{2.5}$ and PM_{10} were measured, possibly because of indoor sources (Fig. 1a–c). Central site outdoor and indoor concentrations of soot, sulfate and especially coarse particles were highest in Athens and next highest in Amsterdam (Fig. 1 d–f).

3.2. Correlation between 24-h average indoor and outdoor concentrations

The distribution of individual correlation coefficients describing the relationship between the 24-h average concentration in the living room and the central site is presented in Fig. 2 and Table 2. The distribution of Spearman and Pearson correlation coefficients was similar. The median correlation was low to moderate for PNs. In Helsinki, Amsterdam and Birmingham, the median correlation for $PM_{2.5}$ was substantially higher. In Athens, the median correlations for PN and $PM_{2.5}$ were similar. Median correlations for coarse particle mass were much lower than for fine particle mass, again with the exception of Athens. Very high correlations in all

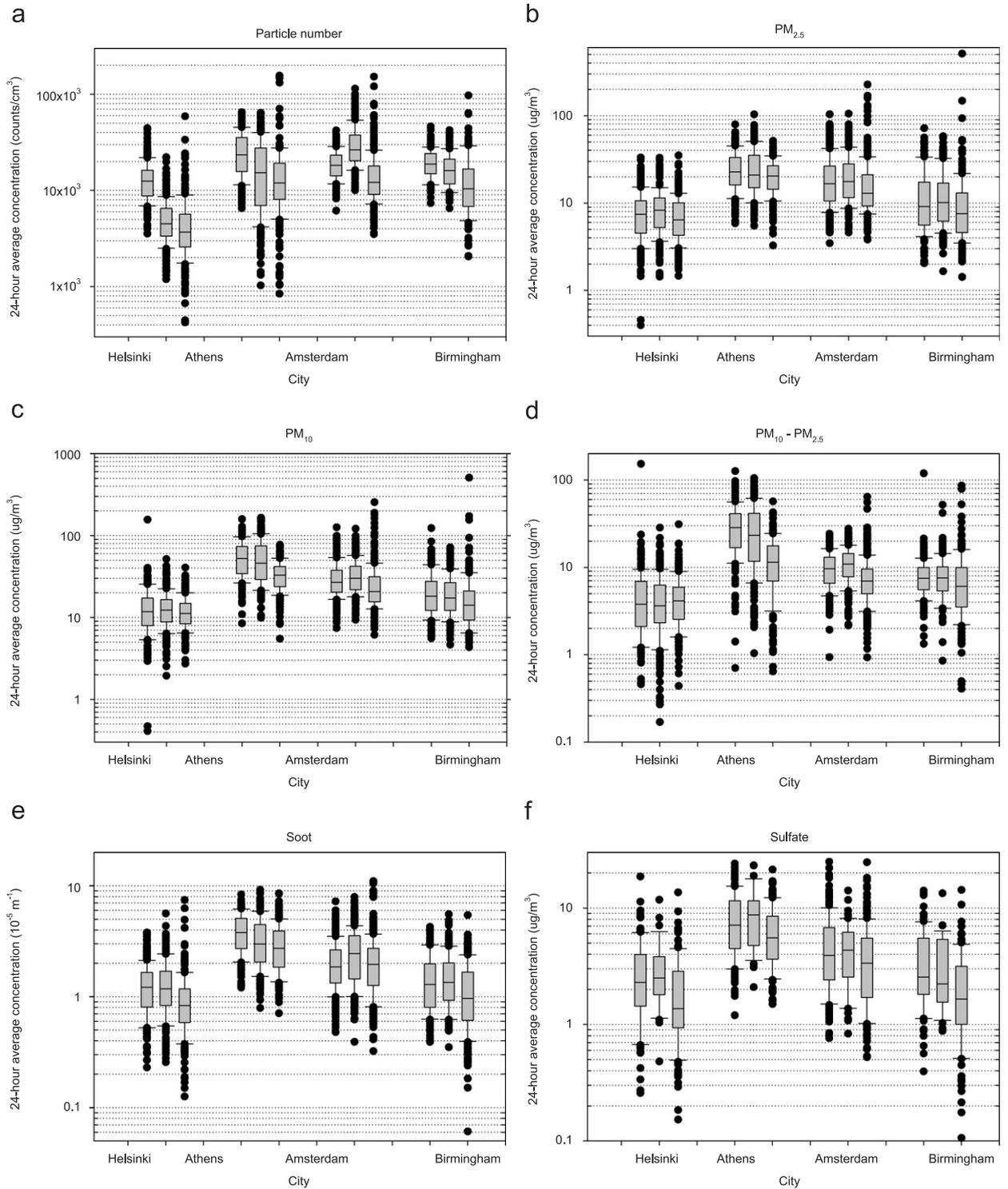


Fig. 1. Boxplots of central site (left), residential-outdoor (middle) and indoor (right) 24-h average concentrations for PN (a), PM_{2.5} (b), PM₁₀ (c), PM₁₀–PM_{2.5} (d), soot (e) and sulfate (f). The centerline of the each box represent the median, the outer lines the 25th and 75th percentiles and the whiskers the 10th and 90th percentile.

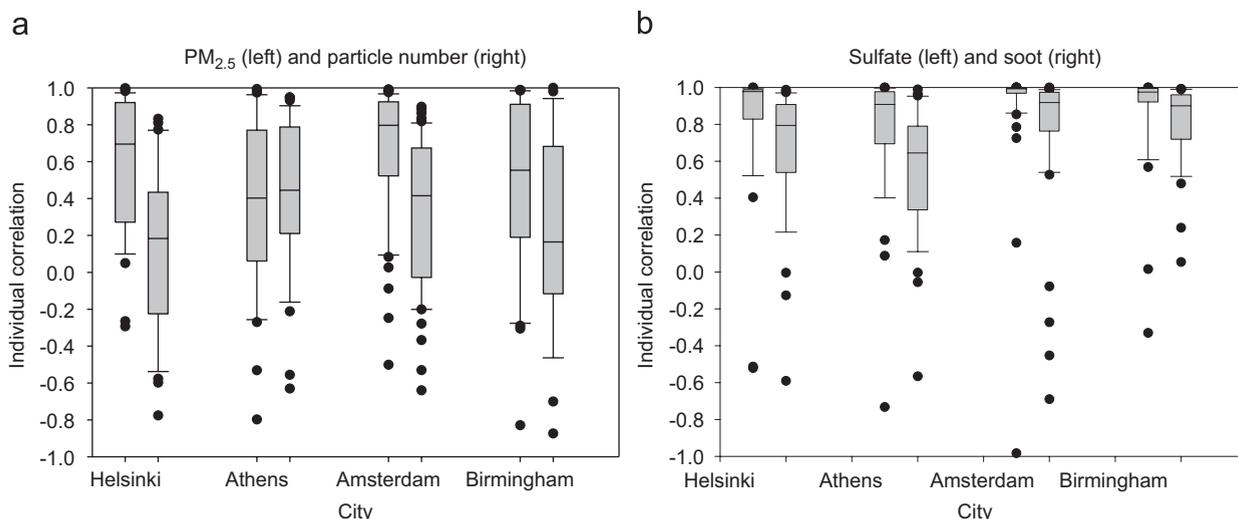


Fig. 2. Distribution of individual correlation coefficients of the relationship between indoor and central site concentrations for (a) particle number and $PM_{2.5}$ and (b) soot and sulfate.

Table 2

Median of individual Pearson correlation coefficients between 24-h average *central site outdoor and indoor* concentrations of particle mass, particle number and composition

	Helsinki	Athens	Amsterdam	Birmingham
PN	0.18	0.45	0.42	0.16
$PM_{2.5}$	0.70	0.40	0.80	0.55
PM_{10}	0.31	0.47	0.77	0.58
$PM_{10}-PM_{2.5}$	0.06	0.39	0.33	0.06
Soot	0.79	0.64	0.92	0.90
Sulfate	0.98	0.91	0.99	0.98

Coefficients in bold were statistically significant ($p < 0.05$) in signed rank test.

Number of homes included per component ranges from 29 to 33 (Helsinki), 30 to 34 (Athens), 49 to 50 (Amsterdam) and 25 to 28 (Birmingham).

Differences in correlation coefficients were statistically significantly different between cities ($p < 0.05$, in non-parametric Kruskal-Wallis test) for $PM_{2.5}$, sulfate, soot, and particle number. For PM_{10} (borderline, $p = 0.07$) and coarse particles differences between cities were non-significant.

cities were found for absorbance and especially sulfate.

The low correlation coefficients for PN and coarse particles were not due a small range in concentrations, related to 1-week consecutive sampling in each home. The median within home range of particle concentrations was typically equal to the overall median concentration. Restricting the analysis to subjects with a range in concentration at the central site that was at least 50% of the home-

specific mean central site concentration did not affect the reported median correlation coefficients. Some of the individual outliers were explained by either a small number of observations or a small range in the central site concentration during the measurement week.

The lower indoor/outdoor correlation found for PN and coarse particle mass compared to $PM_{2.5}$, sulfate and soot could be due to higher spatial variability of ambient air pollution, more losses during infiltration of ultrafine and coarse particles in the home or to more indoor sources for ultrafine and coarse particles.

To address the impact of spatial variation we substituted the residential-outdoor concentration for the concentration at the central site (Table 3). The correlation between indoor and residential outdoor concentrations was higher than between indoor and central site outdoor concentrations, especially for PN. Correlations remained low for coarse particles. This suggests that spatial variation of ambient concentrations was an important factor for PN and less so for $PM_{2.5}$, sulfate and soot, consistent with the analysis of spatial variation reported elsewhere (Puustinen et al., 2007). Various other studies have documented limited spatial variability and high correlations of concentrations measured at multiple sites in an urban area for $PM_{2.5}$ and sulfate (Monn, 2001). Compared to fine particles, PN concentrations are more affected by local sources such as motorized traffic (Sioutas et al., 2005). Consistently, in the few available

studies correlations of PN between multiple sites in an urban area were moderately high (Buzorius et al., 1999; Aalto et al., 2005).

Indoor sources could explain the lower correlations for PN and coarse particles, as various studies have reported that in non-smoking homes, there are more significant sources for ultrafine and coarse particles than for fine particles (Abt et al., 2000a, b; Long et al., 2001; Pekkanen and Kulmala 2004). In the selection of homes, we attempted to minimize the impact of indoor sources, especially smoking.

Table 3

Median of individual Pearson correlation coefficients between 24-h average residential-outdoor and indoor concentrations of particle mass, particle number and composition

	Helsinki	Athens	Amsterdam	Birmingham
PN	0.41	0.80	0.58	0.50
PM _{2.5}	0.74	0.63	0.85	0.35
PM ₁₀	0.57	0.68	0.72	0.52
PM ₁₀ –PM _{2.5}	0.26	0.39	0.26	0.10
Soot	0.96	0.88	0.96	0.93

Coefficients in bold, statistically significant ($p < 0.05$) in signed rank test.

Sulfate not analyzed on residential-outdoor PM_{2.5} samples.

Number of homes included per component ranges from 34 to 36 (Helsinki), 30 to 34 (Athens), 47 to 49 (Amsterdam) and 17 to 27 (Birmingham).

Other sources such as candle burning and cooking/frying could not be avoided under normal living conditions. Excluding the few smoking households and those who reported any smoking during the measurement week, did not affect the correlations for any of the pollutants. Excluding subjects with smoking, candle burning and more than 3% of the time frying and 13.5% cooking during the measurement week, also did not affect correlations substantially (Fig. 3a, b). Comparing correlations across cities, it seems unlikely that indoor sources affected the reported correlations substantially, as the city with the highest proportion of gas used for cooking (Amsterdam) had a relatively high indoor-outdoor correlation for PNs. The impact of indoor sources is further evaluated for hourly PN data.

Higher infiltration of ambient particles into the home for fine particles compared to coarse and UFP is another likely contributing factor to the higher correlations observed for fine particles (Abt et al., 2000b; Long et al., 2001; Sarnat-Ebelt et al., 2006). The higher correlations observed for sulfate and soot compared to PM_{2.5} is probably related to lower losses during infiltration, related to the smaller particle size of these components compared to some other components of PM_{2.5} (e.g. the tail of the coarse fraction) and the non-volatile nature (compared to, e.g. ammonium nitrate). This issue is further discussed in the next section.

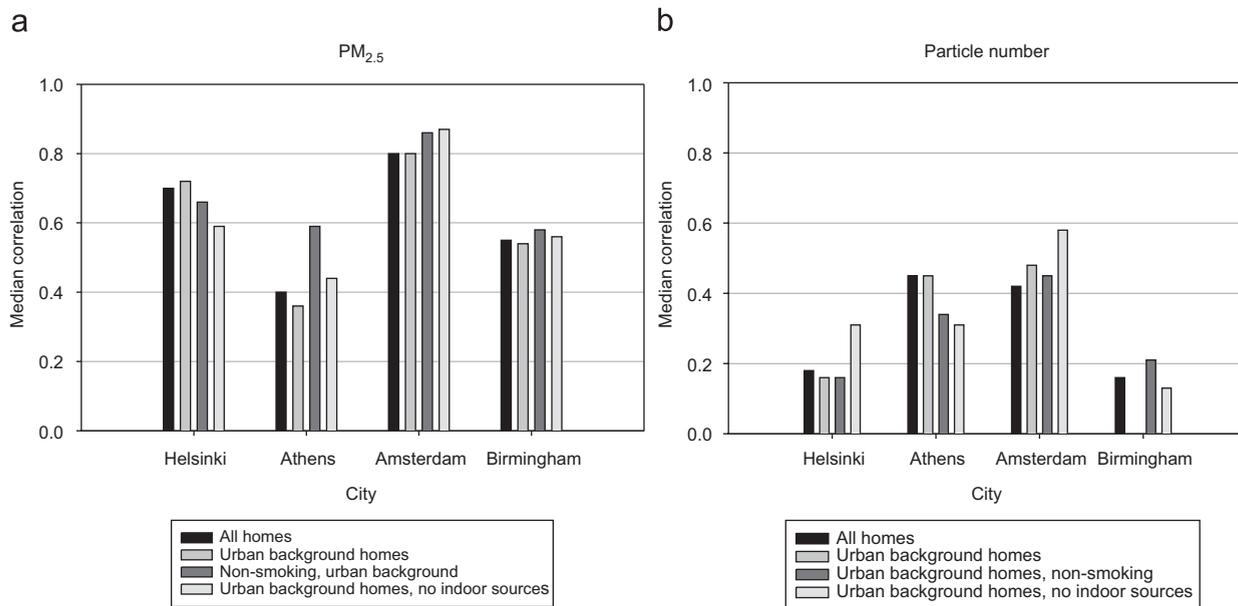


Fig. 3. Median correlation between indoor and central site outdoor concentration in relation to proximity to traffic and indoor sources for (a) PM_{2.5} and (b) particle number.

Further evidence for the impact of ambient air and indoor sources was obtained by evaluation of the average diurnal variability of outdoor and indoor PN concentrations (Fig. 4). Indoor concentrations tracked the outdoor concentrations well, but at a lower concentration. Both indoor and outdoor concentrations were lowest during nighttime hours and experienced a clear morning peak, probably related to the morning rush hour. Indoor smaller peaks are observed at lunch time and in the evening. These may be related to indoor sources, such as cooking and the evening rush hour. The evening rush hour is less pronounced in the outdoor measurements than the morning rush hour.

Previous studies have assessed the correlation between indoor concentrations and outdoor concentrations measured directly outside the home/building. In a study in nine non-smoking Boston homes the Spearman correlations between home indoor and outdoor concentrations varied from 0.67 for $PM_{0.02-0.1}$, 0.90 for $PM_{0.1-0.5}$, 0.83 for $PM_{0.7-2.5}$ and 0.52 for $PM_{2.5-10}$ (Abt et al., 2000b). Indoor concentrations measured in two empty hospital rooms in a hospital in Erfurt were highly correlated to outdoor concentrations measured nearby for $PM_{2.5}$ ($R = 0.88$), soot ($R = 0.91$) and PN ($R = 0.64$) (Cyrus et al., 2004). Indoor PN concentrations were highly correlated to outdoor PN concentrations in an office in Helsinki (Koponen et al., 2001). In a schoolroom in Utah, indoor and outdoor PN concentrations were highly correlated ($R^2 = 0.69$). A study in two retirement homes in the Los Angeles basin found indoor–outdoor correla-

tions of hourly PN data between 0.34 and 0.57; hourly $PM_{2.5}$ between 0.37 and 0.91 and EC between 0.44 and 0.79 (Polidori et al., 2007). The correlations for $PM_{2.5}$ and soot in Amsterdam and Helsinki agree closely with those from an earlier study in these two cities (Janssen et al., 2000; Brunekreef et al., 2005). In that study, central site–indoor correlations for $PM_{2.5}$ were 0.80 and 0.68 in Amsterdam and Helsinki, respectively. For soot, corresponding correlations were 0.93 and 0.70 (Janssen et al., 2000; Brunekreef et al., 2005). Our study thus adds evidence that when outdoor concentrations are measured near the home, variations in indoor concentrations of PN, fine particle mass and fine particle sulfate and soot follow those in ambient air. Our study adds to previous studies that ambient air pollution measured at a central site (common in epidemiological studies) does not correlate well with indoor PN concentrations.

3.3. Indoor/outdoor regression slopes and intercepts of 24-h average data

The median regression slope for PN was less than for $PM_{2.5}$ in Helsinki and Birmingham and similar in Amsterdam and Athens (Table 4). Higher slopes were found for sulfate and soot for all cities. Very low slopes were found for coarse particles in all cities. For PM_{10} and especially $PM_{2.5}$, regression slopes did not differ substantially between cities, whereas the slope for PN differed from near zero (Helsinki, Birmingham) to 0.43 (Amsterdam). All intercepts were significant with the exception of

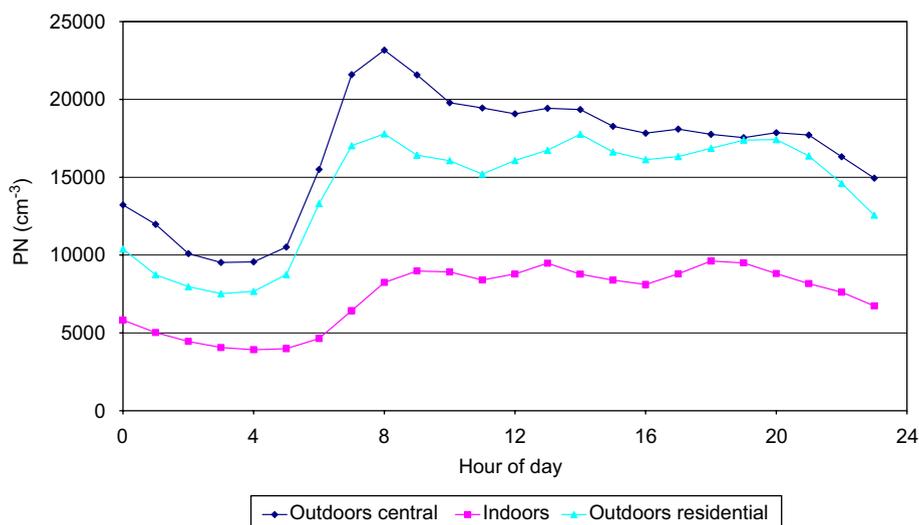


Fig. 4. Diurnal variation of particle number concentrations (median of start hour).

Table 4

Median intercept (α) and regression slope (β) for the relationships between 24-h average indoor and central site outdoor concentrations of particle mass, particle number and composition

	Helsinki		Athens		Amsterdam		Birmingham	
	α	β	α	β	α	β	α	β
PN	3187	0.06	6532	0.25	6099	0.43	6634	0.15
PM _{2.5}	2.6	0.51	10.8	0.30	4.7	0.38	3.3	0.37
PM ₁₀	8.2	0.17	19.7	0.28	7.8	0.41	7.5	0.27
PM ₁₀ –PM _{2.5}	3.9	0.01	7.2	0.17	4.8	0.14	4.8	0.05
Sulfate	0.2	0.59	0.5	0.61	0.0	0.78	0.1	0.61
Soot	0.1	0.58	1.1	0.43	0.1	0.87	0.0	0.66

All intercepts significant with the exception of sulfate in Amsterdam and Birmingham and soot in Birmingham. Units intercept $\mu\text{g m}^{-3}$ for PM₁₀, PM_{2.5} and PM₁₀–PM_{2.5} and sulfate; 10^{-5} m^{-1} for soot; counts cm^{-3} for PN.

sulfate (Amsterdam, Birmingham) and absorbance (Birmingham), pointing to the presence of indoor sources. The intercept was high relative to the overall mean indoor concentrations for PN, PM₁₀ and coarse particles. The intercept was lowest for sulfate and soot.

Our study adds evidence that particle size and composition affect infiltration of ambient particles in the home. Some previous studies have reported that infiltration of coarse and UFP is less than infiltration of fine particles (Abt et al., 2000a; Long et al., 2001; Sarnat-Ebelt et al., 2006). In nine non-smoking Boston homes, the average IF was 0.74 for PM_{2.5} and 0.28 for coarse particles. For particles from 0.02 to 0.1 μm , the IF ranged from 0.5 to 0.7 (Long et al., 2000). For sulfur the average IF was 0.72, virtually the same as for PM_{2.5} (Sarnat et al., 2002). Median indoor/outdoor concentration ratios in two empty hospital rooms in Erfurt (Germany) ranged from 0.79 for PM_{2.5} to 0.43 for PN; for soot the median I/O ratio was 0.53 (Cyrus et al., 2004). Indoor PN concentrations in an office in Helsinki were only about 10% of the corresponding outdoor concentration (Koponen et al., 2001). In a school-room in Utah, the regression slope of indoor and outdoor PN concentrations was 0.14 (Patterson and Eatough, 2000). In two Los Angeles retirement homes, the infiltration factors for PM_{2.5} (0.38–0.52) and PN (0.46–0.78) were similar (Polidori et al., 2007). In 17 Los Angeles homes, the infiltration factor for PN and PM_{2.5} were similar as well, ranging from ~ 0.5 to ~ 0.7 (Sarnat-Ebelt et al., 2006). The infiltration factor was 0.75 for particles between 0.08 and 0.3 μm , 0.50 for the smallest

particles (0.02–0.03 μm) and 0.17 for the largest particles (5–10 μm). Particle removal upon infiltration in the home may occur through diffusion, impaction and gravitational settling. Diffusion is probably important for the smallest particles; gravitational settling for the largest particles (Sarnat-Ebelt et al., 2006; Long et al., 2000). For the accumulation mode particles both mechanisms are less important. The city-average indoor–outdoor slopes for PN in our study ranged from 0.19 to 0.42 (Table 5). The PN infiltration in our study is thus relatively low, probably related to higher losses during infiltration compared to Los Angeles homes or lower air exchange.

Our study shows that infiltration of particles depends on the composition of particles. Infiltration factors were higher for sulfate and soot than for PM_{2.5}, similar to observations in Boston homes (Sarnat et al., 2002), California homes (Sarnat-Ebelt et al., 2006), California retirement homes (Polidori et al., 2007), seven Birmingham homes (Jones et al., 2000) and homes in four European cities (Hänninen et al., 2004). A study in Hong-Kong found regression slopes of about 0.4 for PM_{2.5}, EC and OC in the two mechanically ventilated buildings and 0.8–1.0 in the naturally ventilated homes (Ho et al., 2004). The infiltration for sulfate and soot may be higher because these components are concentrated in smaller particles and are non-volatile. Sulfate and soot are typically concentrated in sub-micron particles (Putaud et al., 2004; Sarnat-Ebelt et al., 2006), whereas PM_{2.5} contains larger particles as well that have lower infiltration factors (Long et al., 2000; Sarnat-Ebelt et al., 2006). Several studies have found that infiltration in the home may result in

Table 5

Median intercept (α) and regression slope (β) for the relationships between 24-h average indoor and residential outdoor concentrations of particle mass, particle number and composition

	Helsinki		Athens		Amsterdam		Birmingham	
	α	β	α	β	α	β	α	β
PN	1731	0.42	2683	0.42	7220	0.19	3745	0.22
PM _{2.5}	2.4	0.48	11.4	0.42	4.7	0.39	4.5	0.34
PM ₁₀	6.4	0.44	16.4	0.38	8.2	0.35	6.0	0.26
PM ₁₀ –PM _{2.5}	3.2	0.14	6.6	0.16	5.1	0.11	4.6	0.13
Soot	0.1	0.63	0.2	0.84	0.1	0.78	0.0	0.71

Sulfate not measured on residential-outdoor filters. All intercepts significant with the exception of soot in Amsterdam and Birmingham. Units intercept $\mu\text{g m}^{-3}$ for PM₁₀, PM_{2.5} and PM₁₀–PM_{2.5} and sulfate; 10^{-5} m^{-1} for soot; counts cm^{-3} for PN.

volatilization of volatile components such as ammonium nitrate (Sarnat-Ebelt et al., 2006; Lunden et al., 2003a; Lunden et al., 2003b). Infiltration factors of 0.2 were found in California homes for particulate nitrate (Sarnat-Ebelt et al., 2006). The equilibrium between ammonium nitrate and the gaseous components ammonia and nitric acid is shifted towards the gaseous components because of temperature and relative humidity changes (Lunden et al., 2003a, b). Moreover nitric acid is removed efficiently by deposition on walls in homes, further contributing to the dissociation of ammonium nitrate (Lunden et al., 2003b). This mechanism may have been important in our study as well. Median nitrate concentrations measured at the central site were 0.4, 1.1, 4.3 and $1.6 \mu\text{g m}^{-3}$ in Helsinki, Athens, Amsterdam and Birmingham. For Birmingham and Amsterdam, this agrees well with the reported average nitrate contribution to $\text{PM}_{2.5}$ of 17% for European urban background locations (Putaud et al., 2004). The 17% contribution predicts nitrate concentrations of 1.3, 3.8, 2.8 and $1.5 \mu\text{g m}^{-3}$ in Helsinki, Athens, Amsterdam and Birmingham. Indoor and central site outdoor concentrations were highly correlated in Helsinki ($R = 0.70$), Amsterdam ($R = 0.83$) and Birmingham ($R = 0.68$). In Athens the correlation was 0.18. Infiltration was low in all cities (range of median regression slopes was 0.05–0.13). The pattern of high correlation and low infiltration agrees closely with the US studies (Sarnat-Ebelt et al., 2006; Lunden et al., 2003b). The interpretation of our data should take into account that our sampling system was not designed to measure nitrate, since we used one Teflon filter for particle collection only. Volatilization during sampling may have occurred and may have biased concentrations downward.

3.4. Hourly particle number data

Indoor–outdoor correlations and regression slopes calculated from hourly PN data were very similar to the corresponding 24 h correlations and slopes (Table 6) when all hours of the day were evaluated. The highest hourly central site outdoor concentrations were close to 200,000 counts per cm^3 , all observed in the early morning hours in Athens. The highest hourly indoor PN concentrations exceeded 1,000,000 counts per cm^3 and were spread over the four cities. Restricting the correlation analysis to observations with indoor concentrations less than 200,000 counts per cm^3 to limit the impact of indoor sources resulted in similar results.

When the analysis was restricted to the night-time period (0:00–6:00), indoor–outdoor correlations became much stronger, whereas the slopes were less affected. Very high indoor–outdoor correlations were found when measurements inside and directly outside the home were compared during night-time hours.

4. Conclusion

Daily average PN concentrations measured at a central site were poorly to moderately correlated with indoor concentrations in a study of 152 homes in four European cities. In three of the cities, the correlation of $\text{PM}_{2.5}$ at the central site and indoors was substantially higher. Very high indoor–outdoor correlations were found for fine particle sulfate and soot. People generally spend a large fraction of their time indoors (Robinson and Nelson, 1995; Klepeis et al., 2001). Assuming therefore that the indoor environment is a relevant proxy for the personal exposure, the lower central site outdoor–indoor

Table 6

Median of individual Pearson correlation coefficients and regression slopes for hourly outdoor PN concentrations at the central site and directly outside the study homes versus indoor concentrations

Outdoor concentration	Time period	Characteristic	Helsinki	Athens	Amsterdam	Birmingham
Central site	All hours	Correlation	0.14	0.27	0.27	0.15
Central site	Night-time	Correlation	0.47	0.55	0.59	0.51
Central site	All hours	Slope	0.06	0.14	0.41	0.22
Central site	Night-time	Slope	0.11	0.15	0.33	0.09
Outside home	All hours	Correlation	0.37	0.62	0.27	0.25
Outside home	Night-time	Correlation	0.87	0.85	0.74	0.64
Outside home	All hours	Slope	0.67	0.48	0.21	0.23
Outside home	Night-time	Slope	0.41	0.48	0.25	0.21

Note: All coefficients statistically significant ($p < 0.05$) in signed rank test. Night-time defined as the 7-h period between midnight and 7:00 a.m.

correlation for PN than for PM_{2.5}, and especially sulfate and soot, suggests that more measurement error occurs for PNs than for fine particle components. Using a central site for exposure assessment for epidemiological time series studies would therefore result in less accurate estimation of the health effect attributable to PNs than to fine particle mass, sulfate and soot. The observation that in Athens, indoor/outdoor correlations were similar for PN and PM_{2.5}, suggests the need for local validation studies for epidemiological studies.

This assessment is correct when we assume that UFP have the same health effect independent of its source/composition. If UFP from ambient and non-ambient sources have different health effects and we are mainly interested in ambient UFP, we should have measured indoor ultrafine concentration of outdoor origin and assessed its correlation with outdoor levels. This is currently not possible, but the analysis of one-hour PN data during night-time hours suggests that this correlation may be substantially higher than for periods with indoor sources.

Residential-outdoor concentrations were better correlated with indoor concentrations than central site outdoor concentrations, especially for PNs. Measurement or modeling of concentrations near the home may help reduce measurement error in epidemiological studies.

Infiltration factors were lower for PN (range 0.06–0.43 across the cities) than for PM_{2.5} (range 0.30–0.51 across the cities) and particularly sulfate (range 0.59–0.78) and soot (range 0.43–0.87). The implication is that the composition of ambient fine particles that infiltrate and remain airborne in homes differs from the composition of fine particles measured at a central site.

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