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Spatial variation of particle number and mass over four European cities

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Abstract

The number of ultrafine particles may be a more health relevant characteristic of ambient particulate matter than the conventionally measured mass. Epidemiological time series studies typically use a central site to characterize human exposure to outdoor air pollution. There is currently very limited information how well measurements at a central site reflect temporal and spatial variation across an urban area for particle number concentrations (PNC).

The main objective of the study was to assess the spatial variation of PNC compared to the mass concentration of particles with diameter less than 10 or 2.5 μm (PM_{10} and $PM_{2.5}$).

Continuous measurements of PM_{10}, $PM_{2.5}$, PNC and soot concentrations were conducted at a central site during October 2002–March 2004 in four cities spread over Europe (Amsterdam, Athens, Birmingham and Helsinki). The same measurements were conducted directly outside 152 homes spread over the metropolitan areas. Each home was monitored during 1 week. We assessed the temporal correlation and the variability of absolute concentrations.

For all particle indices, including particle number, temporal correlation of 24-h average concentrations was high. The median correlation for PNC per city ranged between 0.67 and 0.76. For $PM_{2.5}$ median correlation ranged between 0.79 and

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0.98. The median correlation for hourly average PNC was lower (range 0.56–0.66). Absolute concentration levels varied substantially more within cities for PNC and coarse particles than for $PM_{2.5}$. Measurements at the central site reflected the temporal variation of 24-h average concentrations for all particle indices at the selected homes across the urban area. A central site could not assess absolute concentrations across the urban areas for particle number.

Keywords: Particle number concentration; $PM_{2.5}$; PM_{10}; Spatial variation; Exposure assessment

1. Introduction

Atmospheric aerosol has been documented to cause reduction of visibility (Finlayson-Pitts and Pitts, 2000) and increased mortality, morbidity and decreased lung function (Pope and Dockery, 2006). Industry and motorized traffic are major anthropogenic sources of particles, both directly and indirectly through the formation of secondary aerosols. In most epidemiological studies, health effects were related to the mass concentration of particles with diameter less than 10 μm (PM_{10}). There is an intense debate in the European Union about future Air Quality Directives for Airborne Particulate Matter. The current directive regulates only PM_{10}, but it is likely that a standard for $PM_{2.5}$ (particles with an aerodynamic diameter smaller than 2.5 μm) will be added, as is the case of the US. There is still considerable uncertainty about which physical and/or chemical characteristics of PM are most important as determinants of health effects (Brunekreef and Holgate, 2002). A number of studies suggest that ultrafine particles (particles with a diameter less than 100 nm) may be particularly important because of their very high numbers relative to their mass, and because they may easily penetrate into the bloodstream leading to systemic effects (Delfino et al., 2005).

Exposure assessment for ultrafine particles is still in its initial stage compared to exposure assessment for PM_{10} and $PM_{2.5}$ (Pekkanen and Kulmala, 2004; Sioutas et al., 2005). Epidemiological time series studies of acute effects of outdoor air pollution have typically estimated exposure of people living in an urban area by measurements at one fixed outdoor site. At present it is largely unknown how well measurements at a central site represent the concentrations in a wider urban area for ultrafine particles. Studies in a range of different countries, have documented that spatial variability of $PM_{2.5}$ is generally small in urban areas (Monn, 2001). If concentrations measured at a central site represent the entire urban area less for particle numbers than for particle mass, health effects assessed for ultrafine particles may be underestimated relative to particle mass due to more misclassification of exposure. This could lead to erroneous conclusions concerning health relevant particle fractions.

A study in Helsinki suggested that the temporal correlation of ultrafine particles measured at four sites in Helsinki was moderate to high (Buzorius et al., 1999). A study conducted in Barcelona, Rome and Stockholm found moderate to high correlations of total particle number count between two sites in each city (Aalto et al., 2005). Spatial variability of fine and ultrafine particles may differ because of different sources and different atmospheric lifetime. In urban environments ultrafine particle concentrations are influenced by motor vehicle emissions (Hussein et al., 2004). Typically, 80% or more of the airborne particle number in the urban air are in the ultrafine size range (Morawska et al., 1998; Shi et al., 2001; Sioutas et al., 2005). Most of the particles emitted by engines are also in the ultrafine range (Kittelson, 1998). Industrial pollutants and long-range transport (LRT) affect aerosol particle number and mass concentrations in urban areas. Accumulation mode particles (>100 nm) can be transported over long distances, whereas the Aitken (20–100 nm) and nucleation (<30 nm) mode particles are transported at most a few hundred kilometres. The coarse fraction of PM_{10} is not normally transported more than a few tens of kilometres. The coarse mode contains mainly particles from local sources (Pakkanen et al., 2001).

The current paper evaluates spatial variability of particulate matter air pollution, characterized by mass ($PM_{2.5}$ and PM_{10}) and particle number concentration (PNC). By specifically linking measurements of particle number to simultaneous measurements of $PM_{2.5}$ and PM_{10}, the study provides a direct comparison of the uncertainties in exposure assessment for particle mass and number concentration. For epidemiological time series studies two aspects are important, namely the temporal correlation of concentrations measured at

different sites in the city and the difference in absolute levels of concentrations measured at different sites in the city. The study was performed within the framework of the EU-funded multi-centre study Relationship between Ultrafine and fine Particulate matter in Indoor and Outdoor air and respiratory Health (RUPIOH), which was designed to assess spatial variability and indoor–outdoor relationships of particulate matter relevant for epidemiological studies. An analysis of indoor–outdoor relationships and health effects is reported separately.

2. Materials and methods

2.1. Study design

The study was conducted in four cities located throughout the European Union, namely, Amsterdam (The Netherlands), Athens (Greece), Birmingham (United Kingdom) and Helsinki (Finland). Particle mass (PM_{10} and $PM_{2.5}$) and number (PNC) and soot concentrations were measured. A central site was selected, where continuous measurements were performed during the entire study period. In each city, additional measurements were made in and directly outside approximately 35 homes of asthma/COPD patients in order to evaluate spatial variability. In Amsterdam, 15 further homes located on the main road network were included with additional local funding. In/near each home, measurements were conducted during 1 week. Because of limited availability of equipment, we could measure only in one home simultaneously. The study period was from October 2002 to March 2004. Standardized questionnaires/forms were used to characterize the sampling sites with respect to location in the city, traffic intensity and street configuration. All partners used the same model of samplers and the same Standard Operating Procedures.

2.2. Study areas and sites

Criteria for the central site were: located central in the study area, not located in an industrial area, preferably in a residential area and an urban background location. An urban background location was defined as not in a street canyon, unless the road carries less than 1000 vehicles day^{-1}, distance more than 5 m to the edge of any road, distance more than 50 m to the edge of road carrying more than 10 000 vehicles day^{-1}, distance more than 100 m to the edge of road carrying more than 25 000 vehicles day^{-1}, distance more than 200 m to the edge of road carrying more than 70 000 vehicles day^{-1} and not within 100 m of small point or area sources (garages, parking lots). The definition of urban background does not exclude that traffic impacts occur, as we especially have little information on quantitative impacts on ultrafine particle number counts.

The entire urban area was eligible as the study area for selecting the homes, as in epidemiological time series studies of daily mortality and air pollution. Homes had to be spread over the entire area. We aimed at selecting 25 homes located in small streets and 10 homes in major streets. If traffic homes were selected, the living room had to be located on ground or first floor, because of documented vertical air pollution gradients in major streets. Measurements near the home had to be at the street side of the home for traffic homes.

In Finland, 37 homes were selected from the Helsinki Metropolitan area (Fig. 1a, Table 1). The Finnish central site was located on the University campus area, an urban background location in the suburbs, about 100 m from a high traffic density road (over 40 000 vehicles per day), about 5 km from Helsinki city centre. The elevation difference between the Finnish central site and the nearest major road was about 10 m. The CPC measurements were done 15 m from the ground and the PM measurements on the roof of the University building, 20 m from the ground.

In Athens 35 homes were selected (Fig. 1b). Athens is located in a valley surrounded by mountains (altitude 457–1412 m). Athens' central site was an urban background site in the city centre, about 30 m from the nearest road with a traffic density of 10 700 vehicles day^{-1}. Central site CPC and PM measurements were conducted on top of the air pollution monitoring station on the Medical School Campus, 4 m from the ground.

In the Netherlands, 50 homes were selected from the Amsterdam metropolitan area (Fig. 1c). Amsterdam's central site was a background site in the city centre, about 100 m from the nearest road with a traffic density of 13 400 vehicles day^{-1}. Central site CPC and PM measurements were conducted on top of a museum, 15 m from the ground.

In the United Kingdom 30 homes were selected from the West-Midlands conurbation (Fig. 1d). Birmingham's central site was an urban background site in the inner city suburbs, about 100 m from the

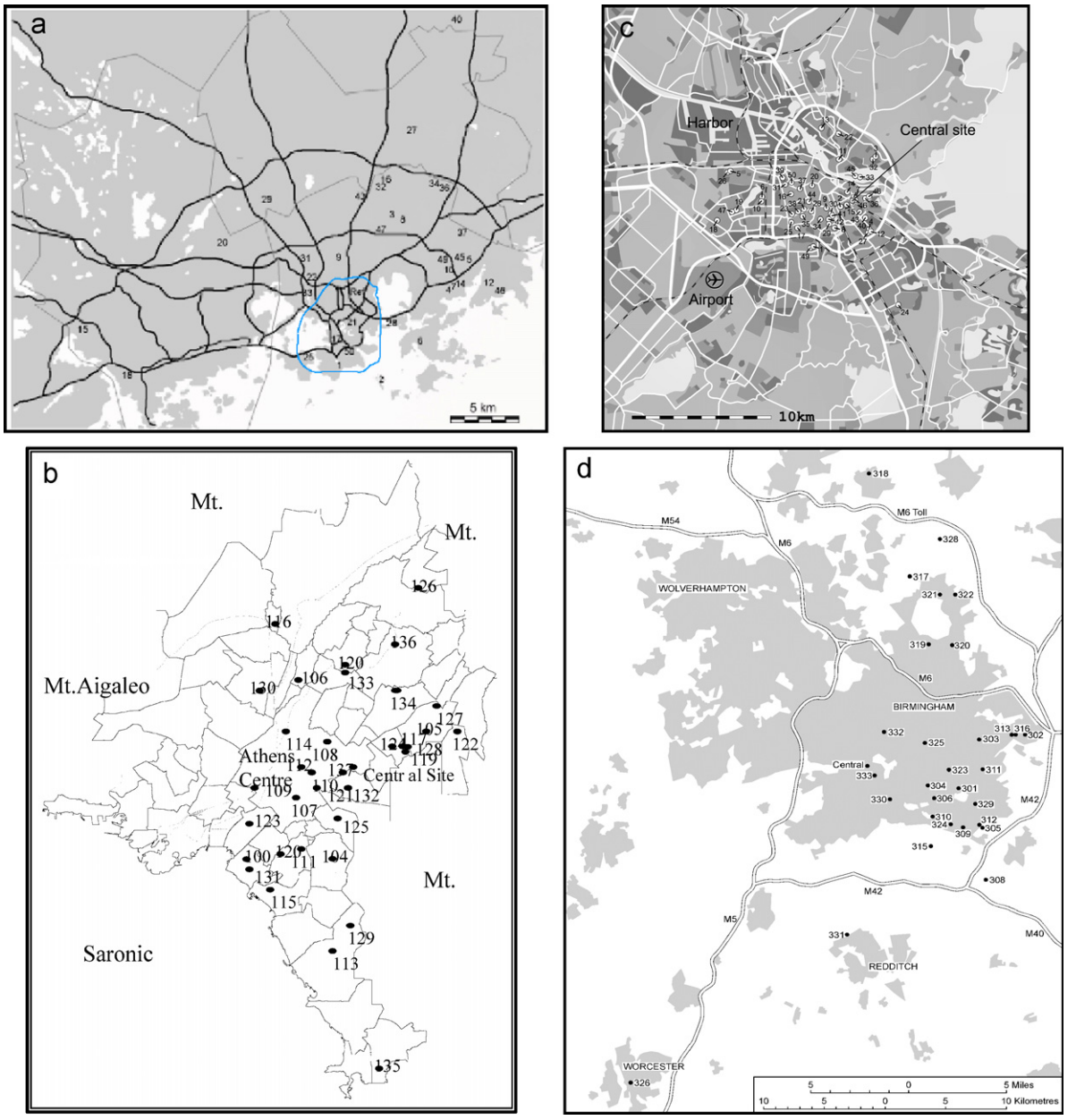

Fig. 1. (a–d) Map of study areas of Helsinki (a), Athens (b), Amsterdam (c) and Birmingham (d). Study homes marked with numbers corresponding home ID and central site marked with Ref or Central.

nearest road. Central site CPC and PM measurements were conducted on the University campus, 1.5 m from the ground.

2.3. Measurement methods

2.3.1. Particle number

Continuous PNCs were measured with condensation particle counters (CPC). In all four cities the same CPC model was used (TSI 3022A, TSI, St. Paul, MN, USA). The CPC 3022A effectively counts particles between 7 nm and 3 μm. Measurements were made according to a common standard operating procedure. The SOP included requirements for the sampling configuration, CPC settings, checking of the proper functioning of the instruments, quality control and data management. To limit particle losses, the inlet had to be constructed from metal, not exceed

Table 1
Description of study areas and study homes

	Helsinki	Athens	Amsterdam	Birmingham
Number of homes	37	35	50	30
Study period	28.10.2002–23.02.2004	23.10.2002–25.03.2004	16.10.2002–23.02.2004	4.11.2002–10.03.2004
Inhabitants metropolitan area (10^6)	1.0	3.2	1.1	2.6
Area (km^2)	745	440	324	902
Population density (km^{-2})	1342	7628	3398	2882
Distance homes to central site (km)[a]	7.6 (2.5–34)	6 (0.7–18.8)	3.3 (0.3–9.5)	7.0 (0.9–29.7)
City centre sites[b]	6 (16%)	9 (26%)	31 (62%)	1 (3%)
Site type[b]				
Background	32 (87%)	22 (63%)	28 (56%)	23 (77%)
Traffic	5 (13%)	13 (37%)	22 (44%)	7 (23%)
Traffic intensity (vehicles per day)[a]	952 (100–8 974)	5046 (100–44 000)	6062 (100–23 446)	2866 (100–19 821)
Canyon street[b]	3 (8%)	11 (31%)	12 (24%)	2 (7%)
Sampling height (m)[a]	3 (0–35)	6 (0–23)	5 (2–25)	2 (2–6)

[a]Median with minimum–maximum distance in brackets.
[b]Number and percentage in parentheses.

150 cm and with gentle bends only. In order to avoid incoming air to condense inside the CPC, a diffusion dryer was installed before the CPC using silica-gel. Condensation created major problems in CPC monitoring in Southern European cities in a previous project (Aalto et al., 2005). A dryer was designed by ECN and distributed to all partners. The dryer consists of a Perspex container filled with sillaca-gel, with an internal metal tube of 20 mm diameter and 30 cm length for the sample flow. If the colour of the sillaca-gel was fading, the cylinder of the dryer had to be replaced (bi-daily checks). After the CPC a box of active carbon to filtrate the gases from the CPC exhaust air was connected. After the active carbon box there was a filter to purify the exhaust air from active carbon dust. Because comparison of concentrations measured at different locations within each city was the main goal of the study, sampling configurations had to be exactly the same within one city. CPC's were operated in the high flow mode ($1.5\,l\,min^{-1}$) to keep residence times in the sampling lines to a minimum. The performance of the CPCs had to be checked at least three times per week. Checking involved mostly reading the performance indicators of the CPC (flow, saturator, condensor, etc. within correct limits). A weekly check had to be performed that included external flow measurements, check of zero counts after the inlet is blocked using a HEPA filter and check of the detector zero. Before the sampling campaign all CPCs had to be calibrated according to the manufacturer's manual. Each month a comparison of the different instruments had to be made at the same location for at least 24 h to document the comparability of the instruments. A comparison was considered satisfactory if instruments agreed within 20%.

For each minute, the average and the minimum and maximum of the available seconds were stored on the computer. We calculated 30 min, hourly and 24-h averages (coinciding with the $PM_{2.5}$ and PM_{10} sampling period) from these 1-min data, if at least 66% of the time period had valid data. To eliminate obvious errors in the data related to the electronics of the instrument, a minute value was excluded if the minimum second value was below 100 particles per cm^3 (usually 0) or if the ratio of the maximum second value and the maximum second value of the next minute exceeded 10. We specifically selected the ratio with the *next* minute to reduce the likelihood that we eliminated true high values related to sources. Only, events caused by sources affecting the site less than 1 min could be filtered out by our procedure. This seems very unlikely given the response time of the instrument and the nature of the monitoring locations. The procedure resulted in deletions of 1.2%, 0.1%, 1.3%, 1.5% of the original central site data in Helsinki, Athens, Amsterdam and Birmingham respectively; 0.2%, 0.1%, 1.1%, 0.83% of residential outdoor data in Helsinki, Athens, Amsterdam and Birmingham, respectively. The vast majority of these deletions were due to deletions related to zero counts: in the UK at the central and outdoor site the percentage of deletions due to the ratio-criterion only was 0.1% and 0.13% compared to the total of 1.5% and 0.8%.

In Helsinki and in Athens two CPC's were used: one in the central site and one for the home measurements, equipped with a sample changer to switch sampling between indoor and outdoor air. In Helsinki, in the field measurements CPC was kept in a wooden box with ventilation. Two metal lines (6 mm radius, approximately 6.5 m length) were taken out of the box, one for outdoor measurements and one for indoor measurements. A sample changer was connected to setup to switch between the lines. The measurement interval was 1 min for each line, out of which 13 s was used to wash out the other line's particle concentration from part of the line and 47 s for sampling the new line. In Athens, approximately the same kind of box with sample changer, dryer and active carbon box with filter was used. Sampling lines were approximately 3 m long. Amsterdam and Birmingham used three different CPC's, one at the central site, one measuring the outdoor air in study sites and one measuring the indoor air in study sites. Sampling lines were approximately 1.5 m long. We calculated the particle penetration efficiency as a function of particle diameter assuming laminar flow conditions using Gormley–Kennedy equations, taking into account the counting efficiency of the CPC-3022. For a 7 nm particle, the penetration efficiency is 84% for a 1.5 m tube and 63% for a 6.5 m tube. However, the counting efficiency of the CPC is only 52%. For a 10 nm particle, penetration efficiencies are 90% and 75% (counting efficiency CPC 71%); for a 20 nm particle, penetration efficiencies are 96% and 89% (counting efficiency CPC 90%). Thus, the tubing introduced small additional losses compared to the counting efficiency of the CPC, with only modest differences between the long and short tubing. For the current study, we were mostly interested in the comparison of concentrations at a central site and near homes within a city and less in absolute levels. We therefore required the configuration used in one city to be the same.

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, which were in all cases traceable to equipment problems in one or two of the CPCs. Field data from these CPCs were deleted from the database. In Finland, intercomparisons were made on only two occasions (February and November 2003), showing good agreement of the two CPCs.

In Finland, no dryer was used at the central site until 22 April 2003. After that date, the ECN dryer was installed. Evaluation of the data series showed that a significant drop of PNC resulted. Further evaluation of the dryer in the laboratory showed significant losses of about 40%. Subsequent field comparisons during 1 week or more of a CPC with the dryer installed and a CPC without the dryer installed showed no losses in the UK, Greece and the Netherlands. The losses in the Finnish CPC were then traced back to a leak in the connections of the dryer to the CPC. PNC after April 2003 were corrected for these losses by multiplying all data by 1.67. The correction factor was derived by regressing the PNC data from the CPC on total particle numbers obtained from a Differential Mobility Particle Sizer (DMPS) operated for another project during the entire duration of RUPIOH at the same site. Before and after April 2003, the ratio of CPC/DMPS counts was 1.12 and 0.67, respectively. Therefore, we corrected PNC data by 1.67. A factor of 1.67 implies 40% losses, consistent with losses found in the laboratory. The consistency with the lab experiments and the high correlation of the PNC data and the DMPS particle number counts with the dryer installed further support the use of this correction factor.

2.3.2. PM_{10} and $PM_{2.5}$

Twenty-four hours average concentrations of PM_{10} and $PM_{2.5}$ were measured with Harvard impactors operating at $10\,l\,min^{-1}$ (Air Diagnostics and Engineering Inc., Naples, ME, USA). Standard operating procedures provided directions for sampling, weighing and reflectance measurements, respectively. These procedures have been described before (Janssen et al., 2000; Brunekreef et al., 2005). Particles were collected on 37-mm 2-μm pore size Teflon filters. The impactors were equipped with a sample changer to measure 3 days continuously, 24 h per impactor. Sample changing was done between 11 and 13 h, preferably at 12 h local time to harmonize sample changing between sites within the same city. Sample volumes were calculated from sampling duration obtained from an elapsed time indicator of the sample changer system and sample flows were measured at the start and at the end of the 24-h sampling period. Flows were measured with calibrated rotameters. Particle mass was determined by gravimetric analysis using analytical microbalances with precision of 1 μg. Teflon filters were conditioned for 48-h in stable temperature

($21 \pm 0.5\,^{\circ}C$) and relative humidity ($35 \pm 5\%$) 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. Absorbance has been shown to be a good surrogate for elemental carbon/soot (Cyrys et al., 2003; Brunekreef et al., 2005). We will further use the term 'soot' for our absorbance measurements.

Quality assurance/control included taking of field blanks, field duplicates and an intercomparison of the weighing and reflectance measurements of the four laboratories.

2.3.3. Data analysis

The data analysis focused on 24-h average concentrations, because this is the main exposure variable in epidemiological time series studies. Moreover this allowed comparison between PNC and PM_{10} and $PM_{2.5}$, the latter available only as 24-h averages.

The correlation between home and central site outdoor measurements was calculated allowing for differences between homes. Correlation coefficients were calculated for each home separately. The main interest is in the median of the individual correlation coefficients. The precision of each individual correlation coefficient is low because of the small number of measurements per home (maximum 7). Both Spearman and Pearson correlation coefficients were calculated.

To assess differences in absolute concentration levels, for each home we calculated the median ratio of the concentrations measured near the home and at the central site. The distribution of the approximately 35 ratios per pollutant was inspected using boxplots. The median of the distribution provided information on the degree in which the central site assesses the average concentration in each city correctly. If the mean differs significantly from unity, the central site provides a biased estimate of the concentration (assuming that the subjects are distributed well over the city). We further assessed the difference between the 25th and 75th percentile of the ratios to describe spatial variation between homes.

3. Results and discussion

3.1. Data quality

Measurements of PM_{10} and $PM_{2.5}$ were successful directly outside 35, 34, 50 and 28 homes in Helsinki, Athens, Amsterdam and Birmingham, respectively. Successful PNC measurements were conducted directly outside 34, 33, 48 and 18 homes in Helsinki, Athens, Amsterdam and Birmingham, respectively. 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 and 5.6, respectively). Reasons for missing data were power failures, programming errors of the PM sampling changer, failure of the computer to store data, malfunctioning of the CPC and termination of the measurements by the inhabitant.

Field blanks and field duplicates indicated that PM measurements were of good quality. Detection limits (DL) for $PM_{2.5}$ and PM_{10} ranged from 1.1 to $4.7\,\mu g\,m^{-3}$ between the four cities. We decided to retain the few values below the DL and not to replace them by a standard value. The relative standard deviation (CV) calculated from field duplicates ranged from 6% to 9% for $PM_{2.5}$ and PM_{10}; 9–24% for coarse particles and 5–10% for soot across the four cities.

3.2. Distribution of particle concentrations

An illustration of the particle number data available for one home is presented in Fig. 2. The figure illustrates the large variability of particle number counts within a week; the diurnal variability with low levels during nighttime in all three monitoring locations. For this specific home, indoor, residential outdoor and central site outdoor concentrations were highly correlated (approximately the 75th percentile of correlations for Helsinki). The outdoor/central site concentration ratio was close to the median ratio for Helsinki. In the remainder of the paper this will be assessed quantitatively.

A description of concentration levels of the various particle indices is presented in Table 2 and Fig. 3 (PNC and $PM_{2.5}$). PNCs at the central and residential sites were lower in Helsinki than in the other cities, consistent with previous findings (Hussein et al., 2004; Wehner and Wiedensohler, 2003). One reason for this could be the lowest population density and traffic density in Helsinki Metropolitan area (Table 1). Differences between the other cities were small. Similar results were obtained by Ruuskanen et al. (2001). Concentration differences between residential sites are difficult to interpret as the distribution of site types was not

equal across cities. $PM_{2.5}$ concentrations were lowest in Helsinki and highest in Athens. This agrees well with previous findings by Ruuskanen et al. (2001). The same trend occurs for PM_{10} and the coarse fraction (PM_{10}–$PM_{2.5}$). Very high coarse particle concentrations occurred in Athens.

3.3. Correlation between central site and residential outdoor concentrations

The distribution of home-specific correlation coefficients of outdoor and central site 24-h average particle concentrations is presented in Fig. 4a–e and Table 3. Pearson and Spearman correlation coefficients were similar. In these analyses only homes with at least three successful simultaneous measurements at the home and central site were included.

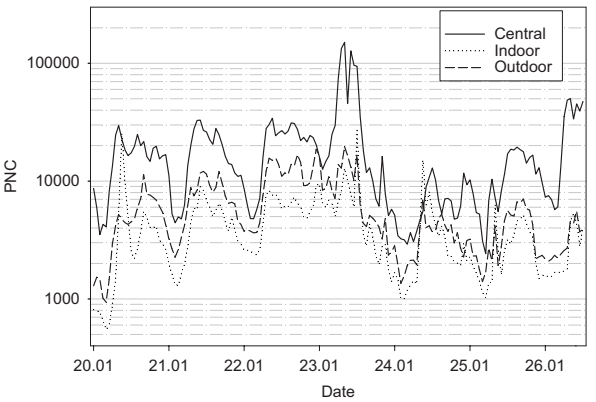

Fig. 2. Example of PNC hourly concentrations in study outdoor site ID 49 for 1 week, in indoor, outdoor and central site. ID 49 was in Helsinki, an urban background site, measured during 20.01–26.01.2004. The residential outdoor/central site correlation was 0.89 and the ratio 0.37.

In all cities, median correlations between central site and residential outdoor concentrations were high to very high for all particle indices, indicating that the temporal variation of particulate matter air pollution near the individual homes was well represented by temporal variation at the central site. Correlations for particle number count and coarse particle mass were lower than for $PM_{2.5}$ and PM_{10}. All correlations tended to be highest in Amsterdam, but differences between cities were small. For particle number, the correlation was slightly higher for the cities with central site measurements at higher sampling height (Amsterdam and Helsinki).

Median correlations with the central site were similar for background and traffic homes (data not shown). For example, the difference in correlation for particle number between traffic and background homes was less than 0.01 and highly statistically insignificant.

The variability of individual correlation coefficients, assessed by the interquartile range (IQR, 75th–25th percentile), was generally higher for coarse particles and particle number than for $PM_{2.5}$. In Athens the IQR for $PM_{2.5}$ and particle number was similar. This suggests that especially for particle number a central site does not represent the temporal variation for all homes well. Note that the correlation coefficient for a specific home should not be interpreted, because of the small number of measurements per home.

Table 2
Median of ambient particulate air pollution concentrations at central and residential sites in four European cities (October 2002–March 2004)

Pollutant	Location	Helsinki	Athens	Amsterdam	Birmingham
PNC (cm^{-3})	Central	12 490	20 276	18 090	18 787
	Residential	4507	15 234	26 346	16 109
$PM_{2.5}$ ($\mu g\,m^{-3}$)	Central	7.5	22.6	16.5	8.7
	Residential	8.3	20.6	17.6	10.2
PM_{10} ($\mu g\,m^{-3}$)	Central	13.1	51.7	26.9	17.1
	Residential	12.3	46.0	29.9	17.2
PM_{10}–$PM_{2.5}$ ($\mu g\,m^{-3}$)	Central	4.5	28.8	9.4	6.8
	Residential	3.6	23.2	10.9	7.6
Soot ($10^{-5}\,m^{-1}$)	Central	1.3	3.5	1.9	1.3
	Residential	1.2	3.0	2.4	1.3

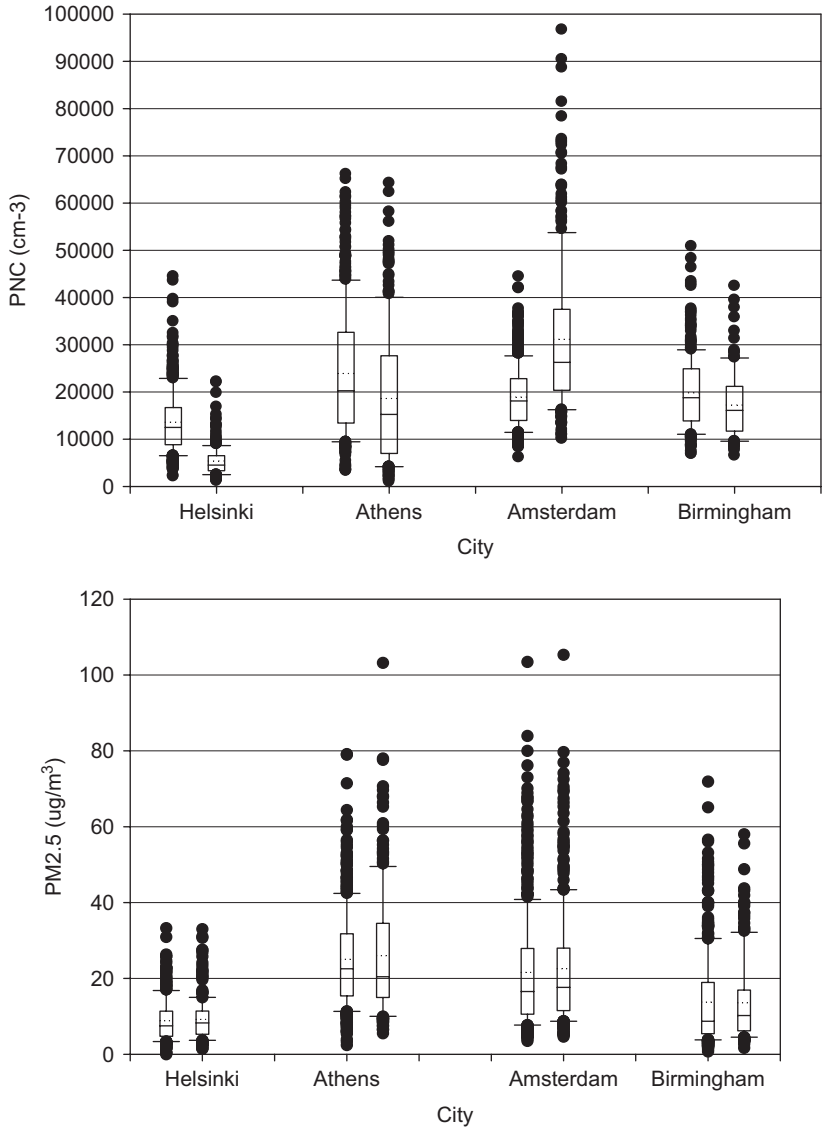

Fig. 3. Distribution of 24-h average central site (left) and residential outdoor (right) PNC and $PM_{2.5}$ concentration. The centre line of the box is the median, the dotted line the mean; the outer lines of the box represent 25th and 75th percentiles, whiskers 10th and 90th percentile.

The very high correlation observed in this study for $PM_{2.5}$ and PM_{10} daily average concentrations between sites spread over the urban area agrees well with early observations for the city of Philadelphia (Burton et al., 1996) and several other studies (Monn, 2001). Factors that contribute are similar weather conditions across the urban area, a high background concentration of regionally transported fine particles and similar impact by urban traffic (a major local source of particles). The somewhat lower correlation for $PM_{2.5}$ in Athens may be due to the larger impact of local sources and local air circulation within the valley that results in a concentration gradient from the centre to suburbs, compared to the other cities where $PM_{2.5}$ is mainly of long-range transport origin. This is in agreement with the larger variation of correlation coefficients between central site and outdoor sites in $PM_{2.5}$ in Athens. The lower correlations for coarse particles are probably due to the more important contribution of local sources, as observed in Philadelphia (Burton et al., 1996). In addition, the lower

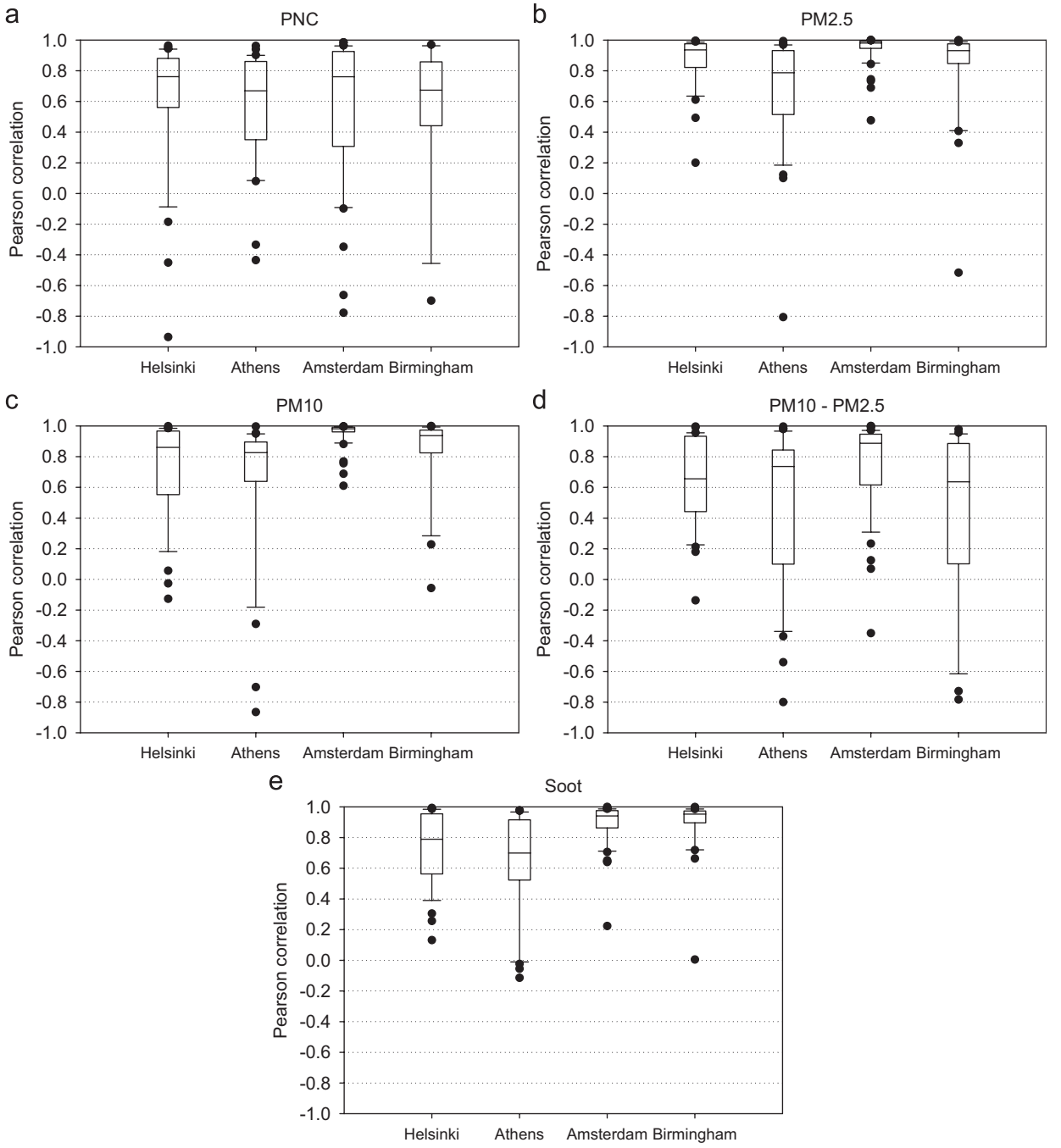

Fig. 4. (a–e) Distribution of individual Pearson correlation coefficients of 24-h average central site and residential outdoor concentrations for different particle indices. The center-line of the box is the median, the outer lines of the box represent 25th and 75th percentiles, whiskers 10th and 90th percentile.

precision of the measurements may have played a role, though this was probably less important as the precision was good for coarse particles as well. E.g. in Amsterdam the measurement precision (expressed as CV) was 11% for coarse particles and 7% for $PM_{2.5}$. This difference probably does not explain the differences in correlation.

The results for particle number agree well with the two previous studies on this topic. A study in Helsinki conducted at four sites found correlation

Table 3
Median and interquartile range (in parentheses) of individual Pearson correlation coefficients between central site and home outdoor 24-h average concentrations for different particle indices[a]

	Helsinki	Athens	Amsterdam	Birmingham
PNC	0.76 (0.56–0.87)	0.67 (0.35–0.86)	0.76 (0.31–0.93)	0.67 (0.44–0.86)
$PM_{2.5}$	0.94 (0.84–0.98)	0.79 (0.51–0.93)	0.98 (0.95–0.99)	0.93 (0.85–0.98)
PM_{10}	0.86 (0.55–0.97)	0.83 (0.65–0.89)	0.98 (0.96–0.99)	0.94 (0.82–0.97)
PM_{10}–$PM_{2.5}$	0.66 (0.46–0.93)	0.74 (0.16–0.84)	0.89 (0.62–0.95)	0.64 (0.17–0.86)
Soot	0.79 (0.56–0.95)	0.70 (0.52–0.92)	0.94 (0.86–0.98)	0.95 (0.90–0.97)

[a]Homes with three or more valid observations.

coefficients of between 0.58 and 0.94 for the comparison of 24-h average concentrations between pairs of sites (Buzorius et al., 1999). The number of measurement days with collocated measurements ranged from 10 to 34 days. Collocated measurements comparing an urban background and a traffic site in three European cities found Pearson correlation coefficients of 24-h average concentrations of 0.67 (Rome), 0.69 (Stockholm) and 0.84 (Barcelona) (Aalto et al., 2005). The number of measurement days with collocated measurements was 293 in Rome, 534 days in Stockholm and 60 in Barcelona. The current study adds a larger number of sites in each area, at the expense of the number of measurements per site. Collectively, our study and previous studies suggest that 24-h average PNC concentrations across the city correlate well with those measured at a central site, though less than for fine particles. The shorter atmospheric lifetime and the larger contribution of local sources of ultrafine particles probably explain the lower correlation compared to fine particles. The occurrence of new particle formation events may have contributed to the observed high correlation for PNC as these would affect the larger urban area (Sioutas et al., 2005). In an earlier study in Birmingham, nucleation processes could be demonstrated on only 3.4% of a total of 232 days spread throughout the year (Alam et al., 2003). Therefore in Birmingham and probably Amsterdam and Helsinki the impact on the observed correlation was probably small.

The continuous PNC measurements allowed us to assess shorter time-scales as well. The correlation between *hourly average* particle number counts measured at the central site and near the home was lower than for the 24-h averages, but still moderately high (Table 4). The moderately high correlation may partly be due to sharing the same diurnal variation. However, the correlation dropped only slightly when the analysis was stratified for daytime and nighttime periods. Buzorius et al. (1999) reported significant correlations between PNC at multiple sites for hourly average concentrations as well.

A limitation of the current study is that due to limited PNC measurement equipment we were not able to measure all homes simultaneously and had to spread the measurements over an approximate 16-months period, including winter and summer seasons. Thus, some of the variability observed between homes may be the result of different weather conditions/seasons of the specific sampling week. Analyses stratified by season (summer versus winter, with winter defined as October–April), showed no significant differences in correlation between homes measured during summer and winter. This does not exclude the impact of more specific weather conditions, e.g. stable atmospheric conditions/regional transport episode. As these conditions vary within the observation period of a home, our design with few measurements per home is not suitable to analyse these impacts in more detail. Studies with fewer sites and more measurements per site are more suitable to address this issue.

3.4. Distribution of ratios of outdoor and central site concentrations

The distribution of home-specific ratios of particle concentrations near the homes and the central site is presented in Table 5 and Fig. 5a–e. To describe the possible contribution of traffic near the residence, all results are presented separately for urban background and traffic homes (Table 5).

While the correlations between central site and residential outdoor concentrations were high, the absolute concentrations of particle number differed substantially between central site and homes spread over the city. In Helsinki and Athens, the central

Table 4
Distribution[a] of individual Pearson correlation coefficients between central site and home outdoor concentrations of hourly average particle number counts

	Helsinki	Athens	Amsterdam	Birmingham
All hours	0.56 (0.37–0.65)	0.58 (0.33–0.66)	0.66 (0.54–0.77)	0.58 (0.41–0.75)
Daytime[b]	0.46 (0.30–0.61)	0.56 (0.19–0.68)	0.60 (0.35–0.74)	0.53 (0.27–0.73)
Nighttime	0.55 (0.39–0.69)	0.56 (0.27–0.65)	0.70 (0.55–0.81)	0.59 (0.39–0.73)

[a]Median and interquartile range in parentheses.
[b]Daytime is the 7–19 h period; nighttime before 7 am and after 19 h.

Table 5
Median and interquartile range (in parentheses) of individual ratios between home outdoor and central site 24-h average concentrations for different particle indices[a]

	Site type	Helsinki	Athens	Amsterdam	Birmingham
PNC	Background	0.38 (0.32–0.50)	0.46 (0.28–0.88)	1.16 (0.97–1.47)	0.85 (0.69–1.24)
	Traffic	0.45 (0.40–0.49)	0.74 (0.58–1.17)	1.93 (1.55–2.93)	0.96 (0.88–1.06)
$PM_{2.5}$	Background	1.02 (0.92–1.20)	0.85 (0.66–1.04)	1.02 (0.98–1.09))	1.08 (0.98–1.19)
	Traffic	1.04 (1.01–1.06)	1.02 (0.88–1.19)	1.17 (1.04–1.31)	1.10 (1.04–1.24)
PM_{10}	Background	1.01 (0.95–1.09)	0.84 (0.66–1.04)	1.03 (0.99–1.09)	1.02 (0.95–1.19)
	Traffic	1.18 (1.18–1.21)	1.22 0.79–1.33)	1.21 (1.08–1.28)	1.06 (0.93–1.09)
Soot	Background	1.12 (0.89–1.26)	0.74 (0.59–0.93)	0.98 (0.88–1.11)	1.01 (0.86–1.05)
	Traffic	1.05 (1.04 1.09)	1.06 (0.75–1.47)	1.44 (1.17–2.07)	1.28 (0.95–1.37)
PM_{10}–$PM_{2.5}$	Background	0.83 (0.66–1.19)	0.88 (0.61–1.13)	1.05 (0.97–1.15)	0.99 (0.93–1.20)
	Traffic	1.30 (0.98–1.53)	1.05 (0.58–1.52)	1.19 (1.07–1.58)	1.04 (0.79–1.10)

[a]Homes with three or more observations.

site overestimated particle concentrations at urban background homes (median home/central site ratio <0.50) and traffic homes (median ratio <0.75). In Amsterdam and Birmingham, the central site resulted in modest over- and underestimation of concentrations near the homes, respectively. The median O/C ratios of traffic homes versus central site were higher in every city than the corresponding O/C ratios for urban background sites.

The differences in O/C ratios for urban background sites observed between Athens/Helsinki on the one hand and Birmingham/Amsterdam on the other hand are difficult to explain. It is unlikely that differences in CPCs explain these large differences, since monthly intercomparisons have been made to ensure that within each city PNCs were comparable within 20%. We cannot exclude a modest impact of nearby major roads on the central site in Helsinki and Athens. In Helsinki the central site was 100 m from a very high traffic density road (more than 40 000 vehicles day^{-1}), but the site was 15 m above ground level. In Athens the site was 30 m from a moderately busy road (10 000 vehicles day^{-1}). It is unlikely that these roads explain the large difference between central site and the homes. We conclude that with current knowledge it is not feasible to select one site to represent absolute concentration levels in the urban area without further evaluation.

The observation of high correlations along with large differences in absolute concentrations for particle numbers agrees closely with measurements at two sites in Stockholm, Rome and Barcelona (Aalto et al., 2005). In that study, the PNC at the traffic site was between 2 and 6 times higher than the concentration at the urban background site.

In all cities, the central site was a good estimate of the median outdoor concentration of $PM_{2.5}$ and PM_{10} at background homes (median ratio close to 1). For the traffic homes, the median home outdoor/central site ratio ranged between 1.02 and 1.22, a modest difference. In addition, the variability in the ratios between homes was

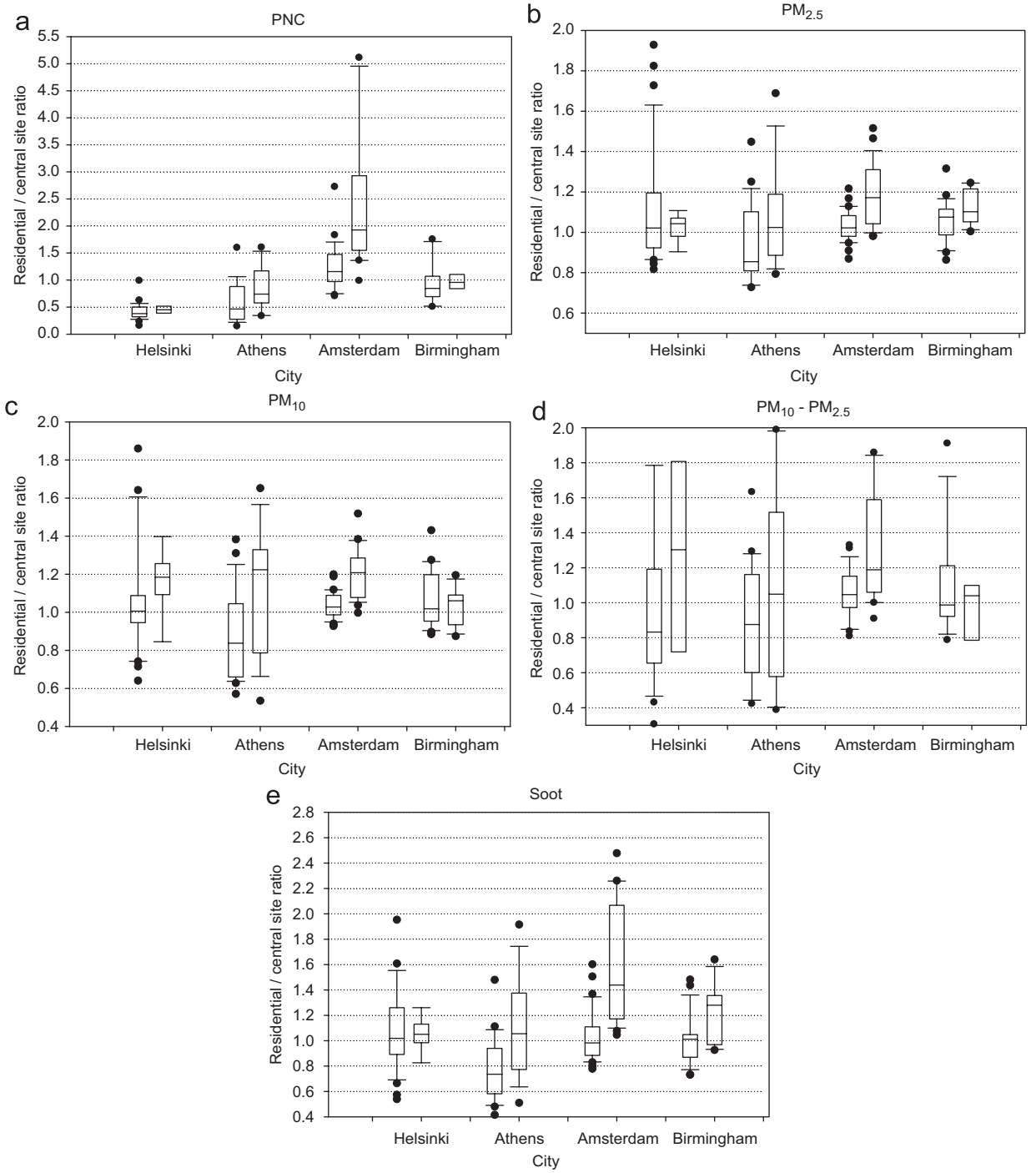

Fig. 5. (a–e) Distribution of the home-specific ratio of the home outdoor/central site 24-h average concentration for urban background (left) and traffic homes (right). The centre-line of the box is the median, the outer lines of the box represent 25th and 75th percentiles, whiskers 10th and 90th percentile.

considerably smaller for $PM_{2.5}$ and PM_{10} than for PNC. This indicates that spatial variability within cities of absolute concentrations was smaller for $PM_{2.5}$ and PM_{10} than for particle number. $PM_{2.5}$ and PM_{10} at one fixed, carefully selected, site in urban environment may thus be representative of

a fairly wide urban area in agreement with previous studies (Gomiscek et al., 2004; Tiitta et al., 2002). The home-specific ratio was more variable in Athens, especially for PM_{10}. These findings indicate that PM_{10} in Athens has more influence from local sources than PM_{10} in the other cities.

In all cities, the central site was a good estimate of the average outdoor concentration of coarse particles at background and traffic homes. The variability between background and traffic homes (IQR, Fig. 5d) was larger than for PM_{10} and $PM_{2.5}$ in all cities, consistent with the notion that the coarse fraction derives mainly from local sources (Pakkanen et al., 2001; Hussein et al., 2004) and has a more limited long-range transport component.

In three cities, the central site was a good estimate of the average outdoor concentration of soot at background homes. In Athens, the concentration at the homes was on average 26% lower than at the central site. At traffic homes, concentrations were higher than at the central site, especially in Amsterdam and Birmingham. The difference between traffic and background homes was larger for soot than for PM_{10} and $PM_{2.5}$.

4. Conclusions

Daily average concentrations measured at a central site reflected temporal variation near homes across urban areas of PNC in every city. One fixed measurement site was not a good approximation for particle number count absolute values over the wider urban area. A fixed central site measuring PNC either over- or underestimated PNC absolute values over the wider urban area. How well the central site represents the area depends strongly where the site is located. Concentrations measured at a central site reflected temporal variation near homes across urban areas of $PM_{2.5}$ and PM_{10} in every city. In addition, only small differences in absolute values between central sites and homes were found, suggesting that one fixed site could represent the wider urban area.

Although the correlation between central site and home outdoor concentrations was higher for $PM_{2.5}$ than for PNC, the difference was not substantial. Using a central site to characterize exposure in epidemiological time series studies, thus does not result in substantial more measurement error for PNC than for $PM_{2.5}$. The larger variation of the absolute concentration level of PNC compared to $PM_{2.5}$ across the cities, suggests that it is virtually impossible to characterize the city-average concentration of PNC with one site. This implies that epidemiological studies assessing health effects related to long-term average exposure between cities, should not rely on one central site, consistent with previous suggestions by Harrison and Deacon (1998), that the number of monitors has to be large in order to cover the spatial variability in cities.

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