TransCom model simulations of hourly atmospheric CO_2

Experimental overview and diurnal cycle results for 2002

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Dependence of Home Outdoor Particulate Mass and Number Concentrations on Residential and Traffic Features in Urban Areas

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IMPLICATIONS

Uncertainties of health risk estimates associated with exposures to PM are mostly due to weak correlations between personal exposures and ambient concentrations, which vary for different fractions of PM. Among other factors, the variability of PM concentration within an urban area is accountable, especially for coarse and ultrafine particles. This study focused on the analysis of spatial variation of fine and coarse PM and ultrafine particles in urban areas. The outcomes of this investigation provide insights on the impact of sampling, home outdoor, and traffic characteristics on the variability of particle mass and number concentrations, which are also critical in developing policies to control atmospheric PM and its sources.

ABSTRACT

The associations between residential outdoor and ambient particle mass, fine particle absorbance, particle number (PN) concentrations, and residential and traffic determinants were investigated in four European urban areas (Helsinki, Athens, Amsterdam, and Birmingham). A total of 152 nonsmoking participants with respiratory diseases, not exposed to occupational pollution, were included in the study, which comprised a 7-day intensive exposure monitoring period of both indoor and home outdoor particle mass and number concentrations. The same pollutants were also continuously measured at ambient fixed sites centrally located to the studied areas (fixed ambient sites). Relationships between concentrations measured directly outside the homes (residential outdoor) and at the

fixed ambient sites were pollutant-specific, with substantial variations among the urban areas. Differences were more pronounced for coarse particles due to resuspension of road dust and PN, which is strongly related to traffic emissions. Less significant outdoor-to-fixed variation for particle mass was observed for Amsterdam and Birmingham, predominantly due to regional secondary aerosol. On the contrary, a strong spatial variation was observed for Athens and to a lesser extent for Helsinki. This was attributed to the overwhelming and time-varied inputs from traffic and other local sources. The location of the residence and traffic volume and distance to street and traffic light were important determinants of residential outdoor particle concentrations. On average, particle mass levels in suburban areas were less than 30% of those measured for residences located in the city center. Residences located less than 10 m from a street experienced 133% higher PN concentrations than residences located further away. Overall, the findings of this multi-city study, indicated that (1) spatial variation was larger for PN than for fine particulate matter (PM) mass and varied between the cities, (2) vehicular emissions in the residential street and location in the center of the city were significant predictors of spatial variation, and (3) the impact of traffic and location in the city was much larger for PN than for fine particle mass.

INTRODUCTION

The impact of urban air pollution on public health is addressed in epidemiological studies in which the risks of respiratory and cardiac diseases and mortality are associated with increased particulate matter (PM) concentrations.[1,2] Many of these studies relied on integrated 24-hr ambient PM concentrations obtained from one (or more) fixed monitoring sites in urban areas. Although most of the exposure studies calculated cross-sectional correlations, instead of more relevant longitudinal analyses, they have demonstrated that correlations between personal and ambient PM_{10} and $PM_{2.5}$ mass concentrations are fairly weak.[3–6,8] The disparity between personal exposure and ambient measurements is partially attributable to the impact of particles generated from indoor sources (e.g., cooking) as well as, ventilation processes[9,10] and individual activity patterns.[7,8,11] The spatial variation of PM also contributes to differences between ambient concentrations and personal exposures. In few cases, the within-city variation for PM less than 2.5 μm in size ($PM_{2.5}$), and to a lesser extent for PM more than 10 μm in size (PM_{10}), is small (on the basis of correlations), suggesting a rather uniform temporal pattern of PM within urban areas[12,13] as compared with recent studies that have demonstrated considerable within-city variations of both PM_{10} and $PM_{2.5}$ in Californian and Canadian urban areas.[14,15] Differences in particle mass between sites were primarily due to contributions from local sources including vehicle emissions, topography, and meteorology.[16–18]

There are only scarce data concerning determinants of exposures to particle number (PN) concentrations and their possible associations with particle mass. Recent studies have shown that ultrafine particles (with aerodynamic diameter <100 nm), which are the predominant contributors to PN concentrations, are important determinants of oxidative stress, inflammatory response, and effects on epithelial cells.[19,20] In those studies, weak correlations between particle mass and number concentrations have been observed in urban areas.[21,22] Furthermore, the spatial distribution revealed significant differences between heavily trafficked and background areas due to diesel exhaust from heavy-duty trucks,[23,24] even in cases when a rather uniform pattern was determined.[25]

A detailed analysis of home outdoor and ambient PM_{10}, $PM_{2.5}$, coarse particle mass ($PM_{10-2.5}$), soot carbon ($PM_{2.5,abs}$.) along with PN concentrations in four European metropolitan areas, Helsinki (Finland), Athens (Greece), Amsterdam (The Netherlands), and Birmingham (United Kingdom) is presented here. The aims of this study are: (1) to determine the degree of spatial uniformity in the particle mass and number concentrations, (2) to identify and assess residential and traffic factors that contribute to the heterogeneity of particle mass and number concentration, and (3) to evaluate their impact on residential outdoor concentrations with respect to measurements at the fixed central sites. The outcomes of this analysis will also allow the determination of the extent to which measurements of a set of particle fractions in a fixed ambient site can be used to describe an urban area and to what degree traffic and other microenvironmental parameters can be used to evaluate the spatial variation of PM.

METHODS AND MATERIALS

Study Design and Sampling

The study was carried out under the framework of the RUPIOH panel study (Relationship between fine and Ultrafine Particulate matter in Indoor and Outdoor air and respiratory Health). A total of 152 (Helsinki, 37; Athens, 35; Amsterdam, 50; Birmingham, 30) subjects with asthma, chronic obstructive pulmonary disease (COPD), or other chronic respiratory diseases were recruited. Subjects were not exposed to occupational pollution and only a few lived in homes with a smoker.[26] The study was divided into two components: (1) a 7-day intensive exposure characterization study that included measurements of PM_{10}, $PM_{2.5}$, and PN concentrations inside and directly outside the home; a 30-min resolution time-activity diary that provided information about indoor sources and ventilation was voluntarily completed by the participants; and measurements of lung function and urine collection to document respiratory health; and (2) a six-month follow-up study in which subjects completed a daily record about respiratory symptoms and medication use. In addition, measurements of ambient particle mass and number concentrations took place in a fixed station located in urban background areas in all centers.[26] The intensive monitoring study was performed sequentially in the various homes (one home monitored per week). The study period was from October 2002 until April 2004. The major advantage of this study is the use of harmonized methods allowing comparison across cities. Similar methods and protocols were used to measure particle mass and number concentration at residences and fixed central sites in all centers,[26,27] thus, differences between cities are unlikely because of measurement-related imprecision.

The details of sampling and measurements of particle mass and number concentrations at fixed, home outdoor, and indoor microenvironments are presented elsewhere.[26] Briefly, 24-hr PM_{10} and $PM_{2.5}$ (from 12:00 p.m. to 12:00 p.m.) were collected on 37-mm Teflon filters (2-μm pore size) using Harvard Impactors operating at 10 L/min.[28] Samplers were installed at least 0.5 m from a vertical surface (e.g., walls). Particle mass was determined by gravimetric analysis using an analytical microbalance with a precision of 1 μg. One-minute PN concentration was continuously measured using a TSI 3022A condensation particle counter equipped with single channel silica gel diffusion dryer to control condensation of water vapor.[26] Absorption coefficients were determined by reflectance measurements of Teflon filters using the ISO 9835 standard "ambient air-determination of a black smoke index" (ISO 9835:1993(E), 1993). Coarse particle mass concentration was estimated as the difference between PM_{10} and $PM_{2.5}$ mass concentrations.

Data Analysis

We calculated the absolute (ΔC) 24-hr paired concentration differences between residential outdoor and fixed central sites. Negative ΔC values indicated higher concentrations at the fixed site as compared with those determined for home outdoor locations. The relative difference (%$\Delta C/Fixed$) was computed as the percentage of the absolute concentration difference to the fixed site concentration. The relative difference also was computed and analyzed for different subgroups of participants on a 24-hr (for particle mass and soot) and hourly (for PN) basis. Median absolute and relative differences provided an indication of systematic differences between the fixed site and outdoors, whereas site-to-site variation was traced by the standard deviation and coefficient of divergence (COD) ratio (see below).

The coefficient of divergence (COD) (eq 1) was used to assess the spatial uniformity of measurements with respect to the concentration levels because correlations between sites only implied a uniform temporal profile during the monitoring period (concentrations decrease or increase simultaneously).[15]

$$\mathrm{COD} = \sqrt{\frac{1}{\mathrm{p}} \cdot \sum_{i=1}^{p} \left(\frac{C_{ij} - C_{ik}}{C_{ij} + C_{ik}}\right)^2}$$

where p is the total number of paired measurements per participant, and C_{ij} and C_{ik} are the measured concentrations in fixed and home outdoor sites on the i-th day, respectively. COD values vary from 0 to 1, with COD values close to unit being indicative of strong spatial variation.

Linear regression analysis was applied to resolve and examine the relationships between home outdoor concentrations, fixed ambient concentrations, and residential/traffic features as follows:

- Univariate regression analysis of the median relative difference as the dependent variable and a subset of residential/traffic characteristics. The following categorized predictive variables were introduced into the models: residence location (center/suburbs), distance from the fixed site (0–10 km, >10 km), residence type (homes/apartments), sampling height (0–5 m, >5 m); traffic-classified residence, and proximity to nearest street (0–10 m, >10 m). The regression coefficients (%) provided a measure of the differences in concentrations between the two categories for each variable.
- Simple regression analysis of daily PM residential outdoor measurements against daily ambient levels (at the fixed site). Regression coefficients (slope and intercept) were obtained for each subject, for each city, and for the entire dataset. Only subjects with at least three successful 24-hr measurements were considered. Despite the within-subject variability, the similarities (and/or differences) between slopes and intercepts provide an indication of the spatial uniformity of PM. The temporal correlation of home outdoor and fixed ambient measurements are presented elsewhere.[27]
- Multivariate regression analysis of home outdoor concentrations against categorized residential/traffic features variables and fixed ambient measurements.[10,18] The variables used in this regression were: residence location (center/suburbs); distance from the fixed site (0–5 km, 5–10 km, >10 km); residence age (before 1980, after 1980); residence type (attached-detached homes/apartments); sampling height (0–3 m, 3–6 m, >6 m); building height (0–9 m, >9 m); traffic-classified residence; proximity to nearest street (0–5 m, 5–10 m, >10 m); proximity to nearest major street and traffic light (0–100 m, 100–500 m, >500 m); and canyon effect and other activities (yes/no). Residences located on streets having uninterrupted buildings on both sides of the street for at least 20 m and the ratio of the building height divided by 2 times the distance between street sides being higher than 1.5 were identified as canyon-type microenvironments.

RESULTS AND DISCUSSION

Residential and Traffic Characteristics

Baseline residential outdoor and traffic characteristics for all centers, collected using standardized forms and questionnaires completed by field operators, are presented in Table 1. About one-third of residences were located in the city center, up to 10 km from the fixed site (the most distant residence at 34 km in Helsinki) (Table 1). Nearly 25% of participants lived in residences constructed after 1980. With the exception of Birmingham, approximately 95% of the residences were apartments in two to seven-floor buildings with a mean building height of approximately 14 m. As a result, sampling was carried out at 5–7 m above the street level. In nearly 65% of studied residences, residential outdoor sampling was carried out just outside of the living rooms with access on the street-side, or both street and back-side of the residence.

Traffic-related data in the vicinity of residences, including distance from the nearest street and major street

Table 1. Residential outdoor and traffic features of residences at four European urban areas.

	Helsinki (n = 37)	Athens (n = 35)	Amsterdam (n = 50)	Birmingham (n = 30)
Residence location				
City Center (n)	6	9	31	1
Distance from the reference site (≤10 km) (n; mean)	30; 9.3	31; 5.3	50; 3.7	21; 8.7
Residence type				
Residences built after 1980 (n)	8	15	9	3
Apartments (n; range of floors)	26; 0–7	33; 0–6	36; 0–7	3; 0–3
Mean (SD) and building height (m)	6.0 (7.5); 14.0 (9.2)	7.0 (5.3); 14.0 (5.5)	5.0 (4.1); 13.0 (7.0)	2.0 (0.9); 8.0 (0.8)
Traffic characteristics				
Traffic homes (n; traffic flow: min–max)	5; 100–8,974	13; 100–44,000	22; 100–23,446	7; 100–19,821
Proximity to nearest street (≤10 m) (n; traffic flow)	16; 1,308	33; 5,311	40; 5,919	13; 100
Proximity to major streets (≤100 m) (n; traffic flow)	7; 34,045	17; 43,703	33; 10,489	2; 19,415
Proximity to traffic light (≤100 m) (n)	6	5	19	2
Canyon effect (n)	3	11	12	2

and traffic flow (vehicles/day) were collected and analyzed. On the basis of these data, residences were classified as:

(1) "Background residence," with low (flow <5000 vehicles/day on the nearest street and there was no street with flow >10,000 vehicles/day within 50 m and 25,000 vehicles/day within 100 m) vehicle load and;
(2) "Traffic residence," with moderate (flow on the nearest street was negligible but there was a major street carrying more than 10,000 vehicles/day within 50 m or 25,000 vehicles/day within 100 m) or high vehicle load (flow on the nearest street >5000 vehicles/day and distance to street <10 m, or flow on the nearest street >10,000 vehicles/day and distance to street <50 m, or flow on the nearest street was >25,000 vehicles/day and distance to street <100 m) (Table 1).

Because of the different construction policies in the urban areas studied, the vast majority of residences in Amsterdam and Athens were located quite close (within 10 m) to streets, compared with less than half of the residences being that close in Helsinki and Birmingham. Consequently, more than 10 residences were located in canyon-type microenvironments in Athens and Amsterdam.

Residential Outdoor-to-Fixed Differences

Table 2 shows the distribution (median and standard deviation) of the absolute (ΔC) and relative differences ($\%\Delta C/Fixed$) between residential outdoor and central fixed site measurements and the COD for each urban area. Negative values of absolute and relative differences indicated that fixed site concentrations were higher than those measured at home outdoor locations at the same time.

Table 2. Absolute (Δ C) and relative ($\%\Delta$ $C/Fixed$) differences (median and SD) of daily concentration of PM_{10}, $PM_{2.5}$, $PM_{10-2.5}$, $PM_{2.5,abs}$. and PN between home outdoor and fixed site measurements, and COD (mean ± SE) for all pollutants in each urban area.

City	PM Metric	Median ΔC	σ (ΔC)	Median $\%\Delta C/Fixed$	σ (% ΔC)	Significance Level	COD
	PM_{10}	0.5	4.7	3.7	34.8	0.161	0.06 ± 0.01
	$PM_{2.5}$	0.7	2.5	8.0	28.6	0.000	0.07 ± 0.02
Helsinki	$PM_{10-2.5}$	−0.2	3.7	−4.1	75.9	0.448	0.50 ± 0.10
	$PM_{2.5,abs}$	0.1	0.6	7.6	45.6	0.030	0.10 ± 0.02
	PN	−4,300	5,100	−44.5	52.8	0.000	0.53 ± 0.04
	PM_{10}	−2.3	25.1	−4.0	43.7	0.182	0.10 ± 0.01
	$PM_{2.5}$	0.2	10.9	0.8	43.6	0.753	0.09 ± 0.01
Athens	$PM_{10-2.5}$	−2.3	20.2	−7.4	65.0	0.107	0.60 ± 0.40
	$PM_{2.5,abs}$	−0.5	1.5	−12.7	38.1	0.000	0.12 ± 0.02
	PN	−9,300	13,100	−34.3	48.3	0.000	0.36 ± 0.06
	PM_{10}	3.1	4.5	10.1	14.7	0.000	0.02 ± 0.01
	$PM_{2.5}$	1.7	3.1	8.1	14.8	0.000	0.02 ± 0.01
Amsterdam	$PM_{10-2.5}$	1.5	3.2	15.0	32.0	0.000	0.07 ± 0.01
	$PM_{2.5,abs}$	0.6	0.9	29.0	43.5	0.000	0.09 ± 0.01
	PN	11,700	16,700	60.5	86.4	0.000	0.22 ± 0.04
	PM_{10}	1.3	3.7	6.4	18.2	0.000	0.02 ± 0.01
	$PM_{2.5}$	0.8	3.3	6.6	27.2	0.008	0.04 ± 0.01
Birmingham	$PM_{10-2.5}$	0.7	3.3	8.9	42.0	0.013	0.20 ± 0.10
	$PM_{2.5,abs}$	0.1	0.4	6.8	27.2	0.054	0.04 ± 0.01
	PN	−1,200	8,200	−6.1	41.7	0.167	0.07 ± 0.01

Notes: Units for metrics: $\mu g/m^3$ for particle mass, $10^{-5}/m$ for absorbance and particles/cm^3 for PN.

High 24-hr PM_{10} and $PM_{10-2.5}$ ΔC values were more commonly associated with increased home outdoor PM_{10} concentration levels (corresponded to $\%\Delta C/Fixed$ values from −7.4 up to 15%; Table 2), whereas no trend was observed for fine particle mass ($\%\Delta C/Fixed$ values from 0.8 to 8.1%; Table 2). This was in agreement with previous studies carried out in areas with low-to-moderate PM levels and high $PM_{2.5}/PM_{10}$ ratios that showed no dependence of ΔC on ambient concentrations.[10,18,29,30] The standard deviation for both absolute and relative differences and the high COD values (from 0.07 to 0.60; Table 2) for $PM_{10-2.5}$ indicated strong site-to-site variations in Athens. This was due to the resuspension of road dust in urban areas, especially during dry periods and without efficient controls.[31] Mechanically generated dust from construction activities for the 2004 Olympic Games may have affected PM levels locally in Athens, although annual PM_{10} concentrations in the region did not vary significantly from 2002 to 2005.[26,32] In Helsinki, road sanding and studded tires during winter and spring may account for the $PM_{10-2.5}$ variability in Helsinki ($\sigma(\%\Delta C)$: 75.9%), although no episodes with high PM_{10} and $PM_{10-2.5}$ concentrations were observed during the monitoring period.

Some indication of the variability caused by the primary $PM_{2.5}$ released from traffic can be obtained by examining the variability of chemically inert fine particle absorbance (an index of elemental carbon [EC]; Table 2). The variability of $PM_{2.5}$ concentrations ($\sigma(\%\Delta C) = 43.6\%$) in Athens exceeded somewhat that computed for $PM_{2.5,abs.}$ ($\sigma(\%\Delta C) = 38.1\%$), providing evidence of the inputs of traffic on fine particles. Other features such as on-street parking and apartment buildings located close to streets with moderate-to-heavy traffic (Table 1) may also have affected fine particle spatial variation in Athens, whereas residences in Helsinki and Birmingham were located far away from heavily trafficked streets. On the other hand, in Helsinki, Amsterdam, and Birmingham, a certain degree of homogeneity due to the influence of regional secondary aerosol on $PM_{2.5}$ concentrations (and differences) was indicated by the low $\%\Delta C/Fixed$ values for $PM_{2.5}$ ($\sigma(\%\Delta C) < 28.6\%$; Table 2), in comparison with those estimated for $PM_{2.5,abs.}$ ($\sigma(\%\Delta C) > 27.2\%$; Table 2). The variability in the four urban areas included in this study was comparable to that observed throughout Europe, with the highest PM concentrations being measured in eastern and southern Europe.[33–35]

Average home outdoor PN concentrations were lower than those measured at the fixed sites in Helsinki (~44%), Athens (~34%), and Birmingham (~6%) with comparable site-to-site variation ($\sigma(\%\Delta C)$: 41.7–52.8%), whereas mean PN concentration at the fixed site was approximately 60% lower than those measured in outdoor home sites in Amsterdam (Table 2). This may be explained by the importance of traffic counts and the distance to the street[23,36] outside residences in suburban areas compared with the fixed site for different centers. In addition, this difference may be related to the difference in percentage of traffic homes in the four cities. The inconsistent degree of spatial homogeneity for PN among centers as indicated by COD values (from 0.07 to 0.53; Table 2), was in accordance with previous studies in Los Angeles,[36] southeastern Texas,[37] central Europe,[38] and Helsinki.[19] Possible contributions of regional transport in Birmingham may result in a rather uniform spatial distribution of PN concentrations levels in the area.[39] On the other hand, elevated traffic emissions in hot-spot locations (such as mixed commercial/residential neighborhoods) may lead to strong spatial variation of PN in Athens and Helsinki.

Effects of Residential and Traffic Characteristics

There are several factors that affect the concentration levels outside the residences, resulting in spatial heterogeneity of concentrations in an urban area. Traffic is considered to be a significant source of primary particles with strong temporal patterns. Meteorological factors also affect the pattern in an urban area by promoting effective convection and mixing. Differences in median relative difference ($\%\Delta C/Fixed$) of particle mass, absorbance, and PN for center versus suburban $[(\%\Delta C/Fixed)_{center}] - [(\%\Delta C/Fixed)_{suburban}]$ and traffic versus background $[(\%\Delta C/Fixed)_{traffic}] - [(\%\Delta C/Fixed)_{background}]$ residences in each city are illustrated in Figure 1, respectively. Low-to-moderate (<20%) differences between center and suburban residences were estimated for particle mass in all four cities. As expected, the suburban $PM_{10-2.5}$ $\%\Delta C/Fixed$ was higher than those computed for residences located in the city center, whereas the opposite was observed for $PM_{2.5}$, $PM_{2.5,abs.}$, and PN. The high relative difference of coarse particle mass for traffic residences in Helsinki and Athens provided further evidence of the significant input of mechanically generated coarse particles from road dust (Figure 1b). Fine particle absorbance and PN values in the city center and traffic residences were higher than those measured in suburban (or nontraffic) sites (Figure 1, a and b).

Median hourly relative differences ($\%\Delta C/Fixed$) of PN in Helsinki, Athens, Amsterdam, and Birmingham are depicted in Figure 2. In addition, Figure 2 shows the effect of residence location (center/suburban) and traffic on PN median hourly $\%\Delta C/Fixed$ in each city. Traffic appeared to be the sole source of these differences in Athens and Amsterdam, with highest differences computed for center and traffic homes during rush hours in the early morning and afternoon. The diurnal pattern for all four urban areas was characteristic of traffic-influenced sites (Figure 2, e–h). A strong bimodal pattern was observed during the morning (6:00 a.m. to 8:00 a.m.) and afternoon (5:00 p.m. to 8:00 p.m.) rush hours in Amsterdam and Birmingham (Figure 2, c and d); whereas in Helsinki and Athens the peak of the diurnal variation was observed around 7:00 a.m. and 12:00 p.m. (Figure 2, a and b). A bimodal distribution was observed in Athens. Traffic appeared to have a larger effect on 1-hr PN $\%\Delta C/Fixed$ values between traffic and nontraffic residences in the morning as compared with afternoon.[38] With respect to traffic load on nearby streets, our results suggest that the location of the residence was accountable for observed home outdoor-to-central fixed site concentration differences for PN. However, the impact of traffic can vary substantially for different urban areas due to local and regional factors such as traffic conditions (speed, fleet), the location of

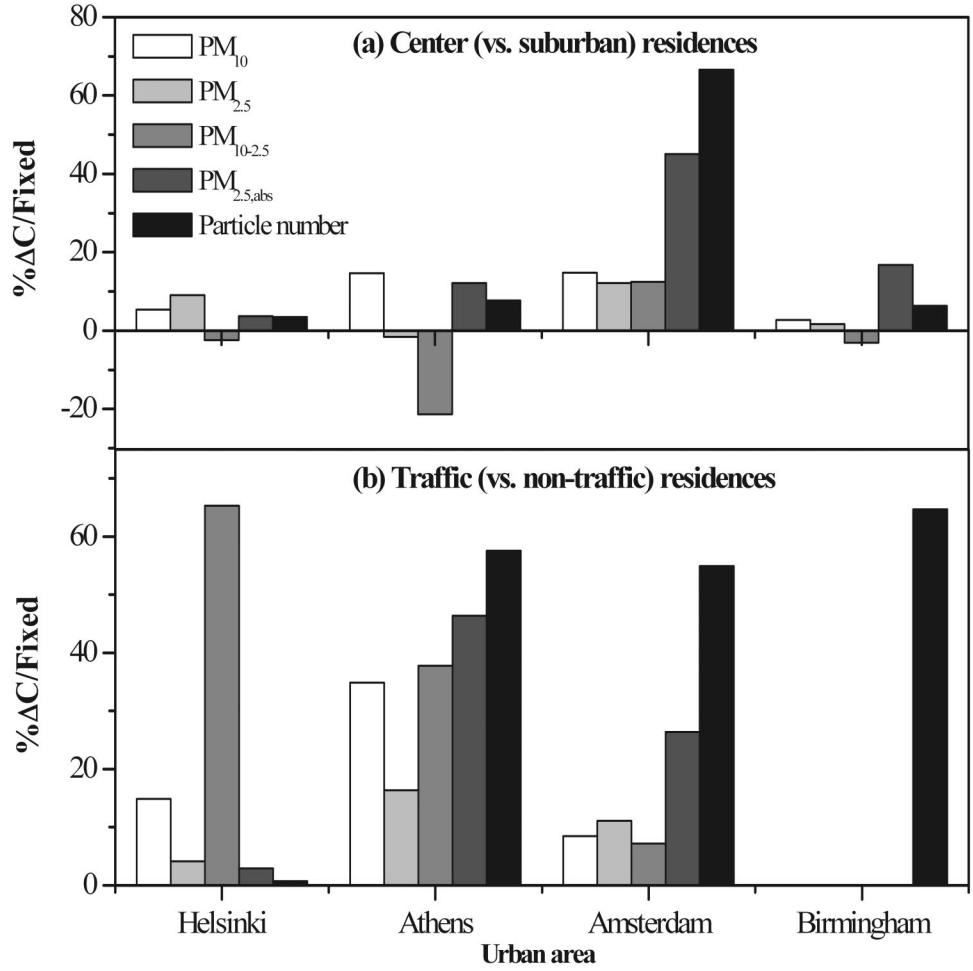

Figure 1. Median difference of daily relative difference of PM_{10}, $PM_{2.5}$, $PM_{10-2.5}$, $PM_{2.5,abs.}$ and PN for different subgroups (a) center vs. suburban and (b) traffic vs. background residences in Helsinki, Athens, Amsterdam, and Birmingham.

residences with respect to roads (e.g., distance from a road, on-road traffic), and climatology.

To further investigate the degree of influence of residential and outdoor characteristics, univariate regression models were applied to explain the concentration difference on a specific day by recorded residential and traffic parameters. Table 3 presents the regression coefficients ± standard error (%) from models with the home-specific median relative difference between residential outdoor and central site ambient concentrations as the dependent variable and the listed variables as independent. Homes located in the city center had between 8 ± 4 and 94 ± 14% higher concentrations than homes in the suburbs (Table 3). A similar profile was also observed for homes located near roads with high traffic load (Table 3). The largest urban/suburban and traffic/nontraffic contrasts were found for PN and $PM_{2.5,abs.}$(>37 ± 7%; Table 3). Traffic load in the vicinity of the residence (within 10 m of the edge of the road) appeared to have a high impact on %Δ*C*/*Fixed* values for particle absorbance and PN concentrations (69 ± 9% and 133 ± 21%, respectively; Table 3). It should be noted that the latter was only calculated for Athens and Amsterdam because there were no homes within 10 m of a major street in Helsinki and Birmingham (Table 1). Other variables not shown in Table 3, such as season of sampling, age, and type of residence did not have an effect on %Δ*C*/*Fixed* values (Table 3).

Fixed-to-Outdoor Relationships

Although regression coefficients between residential outdoor sites and the fixed sites for each subject provide an index of the temporal correlation between the sites (concentrations decrease or increase simultaneously), changes of regression coefficients from subject- to city-specific and single (all subjects/all cities) models were related to the different degree of spatial homogeneity in concentration units (similar concentrations in both sites) for particle mass and absorbance. The median (±standard deviation) of the individual regression coefficients (slopes and intercepts) between central fixed site and home outdoor concentrations are presented in Figure 3 (open squares). Slopes of individual models for particle mass varied from 0.39 to 1.03 (Figure 3, a–c) and median intercepts were lower than 10 $\mu g/m^3$ (Figure 3, f–h). Home outdoor fine particle absorbance ($PM_{2.5,abs.}$) measurements were moderately related to corresponding measurements at the fixed site with median slopes from 0.58 to 1.04 and intercepts lower than 0.70×10^{-5} m^{-1} (Figure 3, d and i). On the contrary, rather poor associations between residential

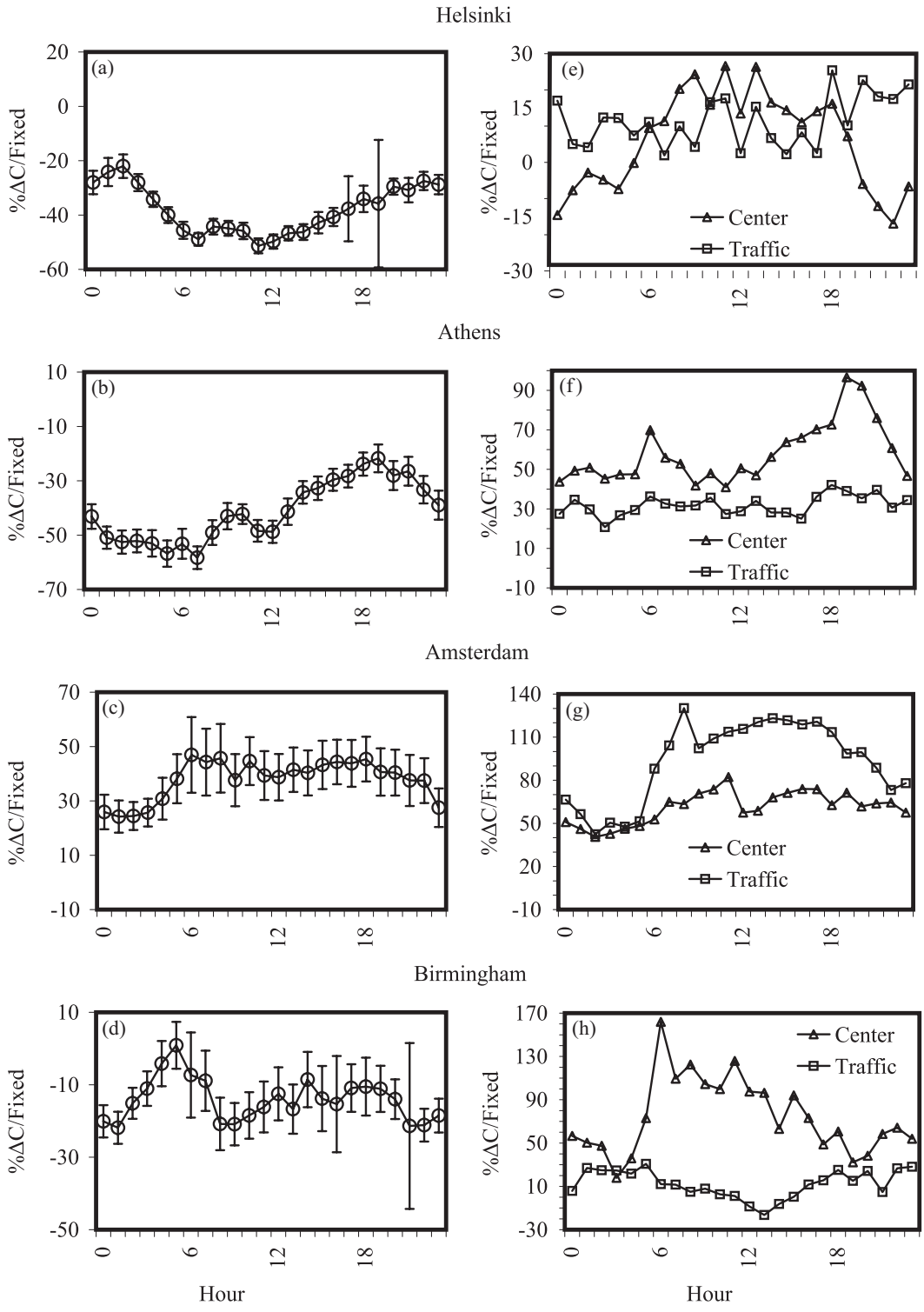

Figure 2. Diurnal variation of the median (±standard deviation) relative difference of PN for all residences, and for center and traffic residences in (a and e) Helsinki, (b and f) Athens, (c and g) Amsterdam, and (d and h) Birmingham.

outdoor and central site number concentrations were observed with subject-specific slopes from 0.36 to 1.22 and intercepts as high as 7000 particles/cm^3 (Figure 3, e and j). For most of the cases, city-specific slopes (black circles in Figure 1) were comparable (within 30%) with subject-specific slopes. A larger variation (from 40% in Athens and Helsinki up to almost 300% in Amsterdam; Figure 3e) was estimated for PN concentrations. With respect to differences among the urban areas, regression parameters obtained from a single model for each parameter (continuous line in Figure 3) for particle mass did not vary significantly (less than 20%) as compared with those computed for each city. As for $PM_{2.5,abs.}$ and PN, estimated coefficients may lead to significant underestimations in Amsterdam (up to 85%; Figure 3, e, i, and j). For all PM fractions, the temporal variation of residential outdoor

Table 3. Effect of home outdoor characteristics on the relative difference between residential outdoor and fixed site concentrations estimated as the median ($\pm\sigma$, %) of the regression coefficients of regression models with relative differences (%Δ *C/Fixed*) of PM_{10}, $PM_{2.5}$, $PM_{10-2.5}$, $PM_{2.5,abs.}$ and PN as dependent variables and the residential/traffic feature as determinants.

Parameter	%PM_{10}	%$PM_{2.5}$	%$PM_{10-2.5}$	%$PM_{2.5,abs.}$	%PN
City center (with suburban residencies)	14 ± 4	8 ± 4	23 ± 7	43 ± 6	94 ± 14
Traffic (with urban background)	11 ± 4	9 ± 3	21 ± 7	37 ± 7	75 ± 15
Proximity to nearest major street (<10 m)	20 ± 6	18 ± 5	30 ± 10	69 ± 9	133 ± 21
Distance to reference site (<5 km)	6 ± 3	4 ± 3	14 ± 6	29 ± 7	74 ± 14
Residence type (apartment vs. single homes)	−5 ± 4	−6 ± 4	−4 ± 8	−14 ± 7	−19 ± 16
Sampling height (<5 m)	−4 ± 4	−1 ± 4	−1 ± 7	−4 ± 8	−10 ± 15

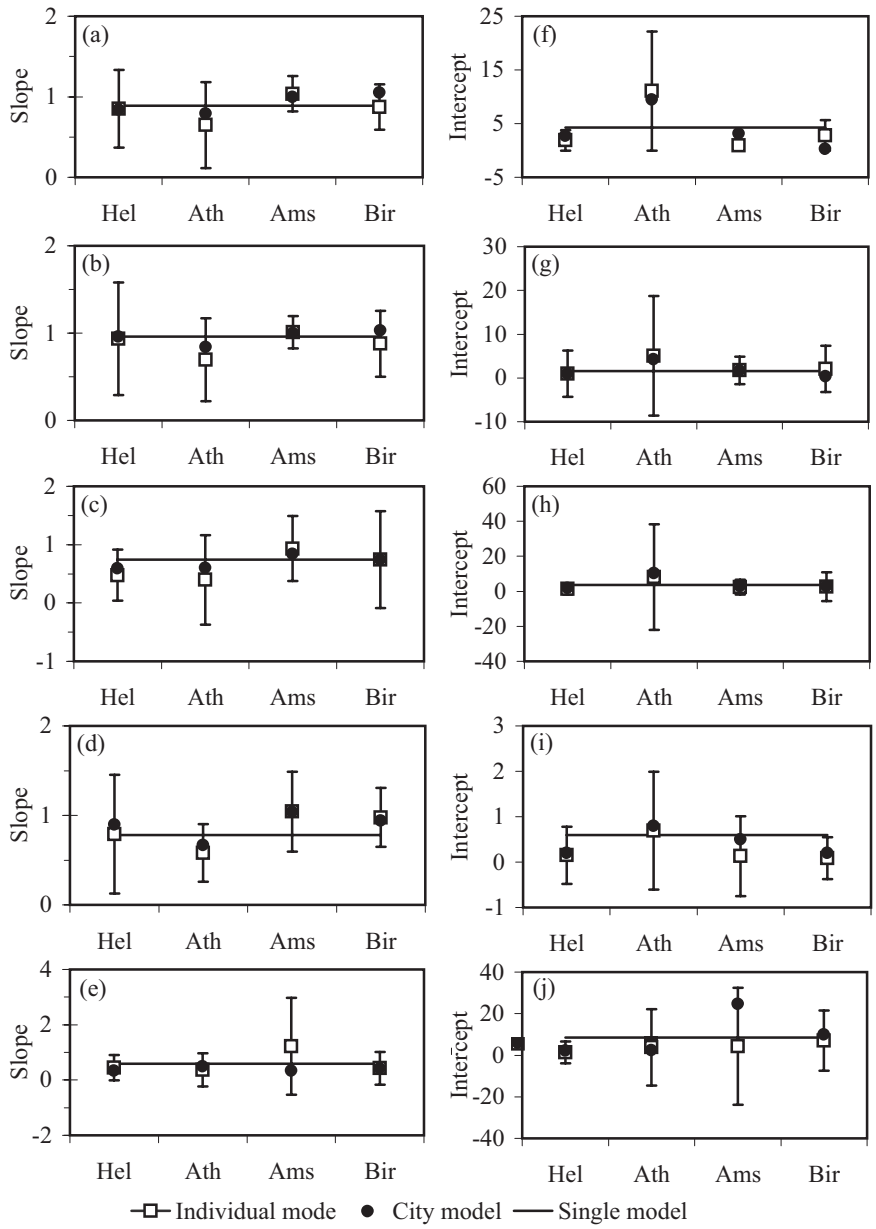

Figure 3. Comparison of regression coefficients (median ± σ) of home outdoor versus fixed site concentrations for (a and f) PM_{10}, (b and g) $PM_{2.5}$, (c and h) $PM_{10-2.5}$, (d and i) $PM_{2.5,abs.}$, and (e and j) PN for subject-specific model (open squares), for city-specific model (black triangles), and a single (all subjects/all cities with city indicators; solid line). Hel = Helsinki; Ath = Athens; Ams = Amsterdam; Bir = Birmingham. Units for metrics: $\mu g/m^3$ for particle mass, 10^{-5}/m for absorbance, and particles/cm^3 for PN.

concentrations was represented quite well by the variability of fixed site concentrations with the highest being measured in Amsterdam as compared with the other urban areas.[27] Correlations for $PM_{10-2.5}$ mass and PN concentrations (0.67–0.76) were slightly lower than for $PM_{2.5}$ and PM_{10} (0.79–0.98).

Determinants of Residential Outdoor Concentrations

Multivariate linear models were used to estimate the impact of fixed site concentrations, and residential and traffic determinants on home outdoor concentrations (Figure 4). In this analysis, applied models accounted for approximately 4–93% of the variation in all cities. All recorded parameters (included in Table 1) were taken into account. For particle mass and fine particle absorbance, fixed ambient site concentrations accounted for a large fraction of home outdoor levels, especially reflecting similar temporal variation within the city. The precision varied across pollutants.

The traffic flow on the nearest street (including the distance to the nearest street) and distance to the nearest traffic light (2.4 $\mu g/m^3$), age (1.1 $\mu g/m^3$), and type (1.5 $\mu g/m^3$) of the home also were important predictors for PM_{10} and $PM_{2.5}$. Seasonal variation, age, and floor (if apartment) of the residence as well as distance from the nearest major street and intersection were insignificant determinants of home outdoor $PM_{2.5}$ mass. For coarse particles, traffic counts and the distance to nearest traffic light explained 1.1 and 1.5 $\mu g/m^3$, respectively. The type of the residence appeared to be a good predictor (1.1 $\mu g/m^3$). Apartment buildings are usually located closer to roads, whereas single-family residences are further away. In addition, the presence of sources of coarse particles in suburban areas (e.g., "dusty" roads) as compared with high traffic in downtown areas could explain the observed different effect of the location of the residence on outdoor measurements. Fine particle absorbance predictors were related to traffic counts and characteristics (distance to nearest street, traffic light) as well as the location of the residence, representing approximately 25% of home outdoor levels. The higher emissions of PM from vehicles at traffic lights (during red-light waiting period and start-up) may also be an important contributor of PM outside of residences.

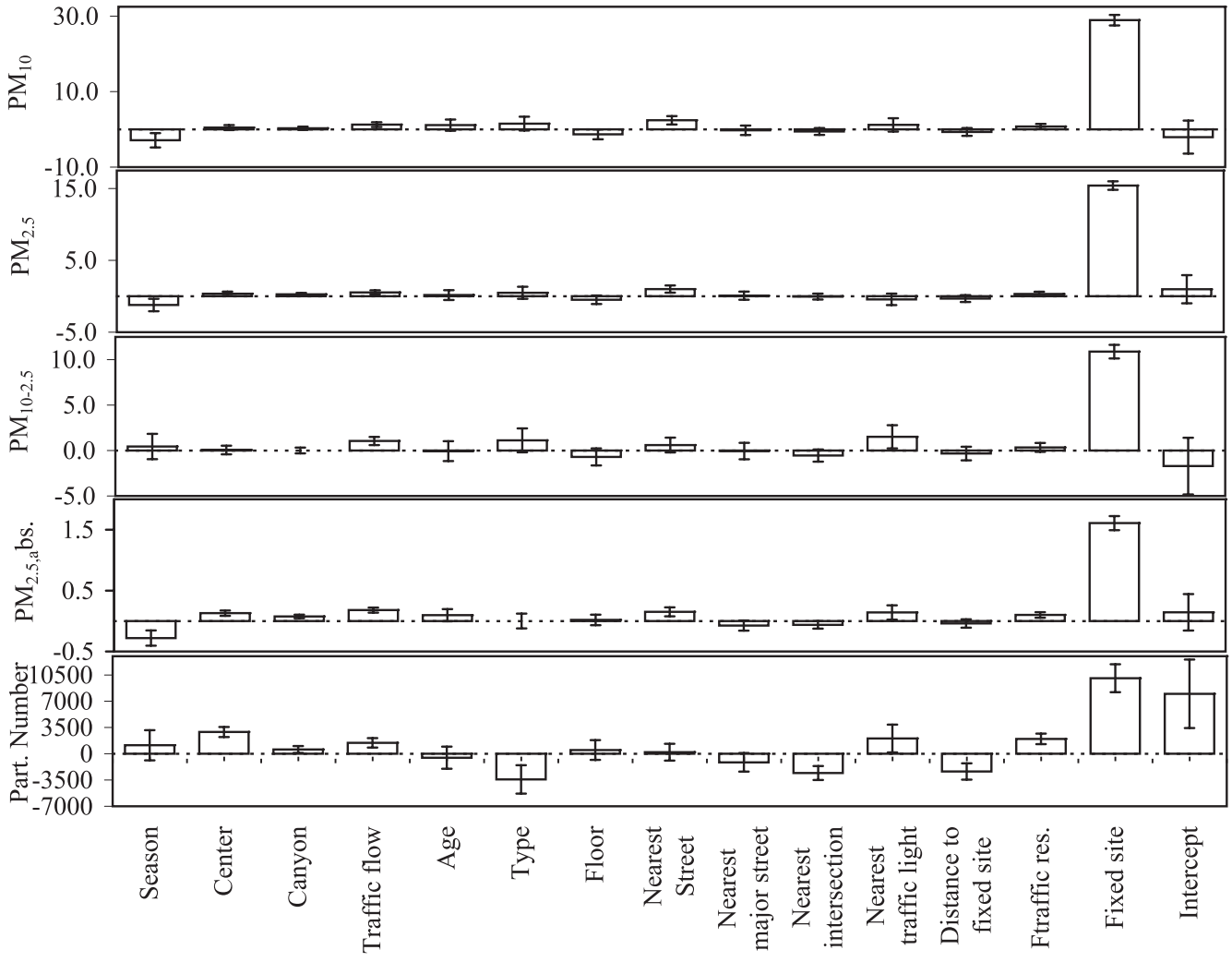

Figure 4. Multiple regression analysis of residential outdoor concentrations for all urban areas. Unit for metrics: $\mu g/m^3$ for particle mass, m^{-1} for absorbance, and particles/cm^3 for PN.

PN concentrations measured at the fixed site accounted for 53% of home outdoor levels. The location of the residence, the distance to the nearest traffic light, and traffic counts were responsible for, on average, 8500 particles/cm^3. Lower particle counts were determined as the distance from the nearest streets and intersections increased and for apartments (as compared with single family residences). A large fraction (40%) of PN was not represented by the fixed ambient measurements and the residential/traffic characteristics recorded in this study. Overall, differences in concentrations between a central fixed site and home outdoor sites spread over the city were larger for PN than for fine particle mass.

CONCLUSIONS

The effects of residential and traffic characteristics on PM concentrations measured outside homes and apartments in relation to central fixed site measurements were investigated in four European urban areas: Helsinki, Athens, Amsterdam, and Birmingham. Differences between the residential outdoor and fixed ambient site concentrations varied substantially for different urban areas and pollutants and correlated with ambient concentration levels. Spatial variability for PM_{10}, $PM_{2.5}$, $PM_{10-2.5}$, $PM_{2.5,abs.}$, and PN in all centers was due to differences in the contribution of local sources of primary PM and transported flux of secondary PM. In addition, meteorological regimes and local topography may also have played an important role by affecting mixing and uniform dispersion.

Spatial variation of PM in an urban area may cause misrepresentation of concentration levels outside of residences (and thus, personal exposures) for epidemiological studies using a single fixed site. This uncertainty varies for different metrics of PM (e.g., particle mass fractions) and depends of the characteristics of the residence. More specifically, concentration differences were related to the residence location (center/suburb) and traffic load near the residence. The diurnal profile of PN concentration differences suggested the significant input of traffic emission in the morning and late afternoon (during rush hours). On the other hand, specific residential characteristics such as the age of residence and sampling/building height did not contribute to the differences between residential outdoor and ambient measurements of particle mass.

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