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Indoor and Outdoor Particle Number and Mass Concentrations in Athens. Sources, Sinks and Variability of Aerosol Parameters

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ABSTRACT

The scope of this work was to characterize PM mass and number concentration at typical residential microenvironments in the centre of Athens and to examine the relative contribution of the indoor and outdoor sources. Three residential flats located in densely populated residential areas were studied, during a warm and cold period of 2002. PM_{10}, PM_2 and black carbon (BC) mass concentrations, as well as ultrafine and accumulation mode particle number size distributions were recorded indoors and outdoors simultaneously. Outdoor concentrations of all size fractions were significant, and indicative of urban sites affected by heavy traffic. Indoor levels were generally lower than the corresponding outdoor ones. Nevertheless, elevated indoor concentrations were recorded, caused by increased ambient air penetration in the indoor microenvironments and/or indoor particle generation. The mean 24-hr indoor PM_{10} concentration at all residences was 35.0 ± 10.7 μg/m^3 during the warm period and 31.8 ± 7.8 μg/m^3 during the cold period. The corresponding PM_2 concentration was 30.1 ± 11.1 μg/m^3 and 27.2 ± 3.6 μg/m^3 during warm and cold periods, respectively. Regression analysis of indoor and outdoor concentration data revealed that indoor BC may be considered mainly of outdoor origin. A large fraction of the outdoor-generated PM_2 and ultrafine and accumulation mode particles also seems to penetrate indoors, causing elevated indoor levels. Regarding indoor particle generation, cooking was the strongest contributor in residential microenvironments.

Keywords: PM_{10}/PM_2; Particle number size distribution; Black carbon; Residential microenvironment; Indoor/outdoor sources.

INTRODUCTION

Airborne particulate matter (PM), and especially fine particles, has been associated with various adverse health effects (Pope, 2000; Pope *et al*., 2002; Pope and Dockery, 2006). Recent toxicological studies have demonstrated that ultrafine particles may have a greater impact on human health than larger particles (Donaldson *et al*., 2002; Ibald-Mulli *et al*., 2002). Given that air quality standards correspond to ambient air, the majority of epidemiological studies attempt to relate health effects with outdoor PM concentrations. Nevertheless, it has been acknowledged that ambient concentration levels may not be indicative of human exposure, since urban populations tend to spend the majority of their time indoors (Brown, 1983).

In this framework, during the last few decades, research interests have turned towards the indoor microenvironments, recognizing their significant contribution to total personal exposure. Considerable focus has been given to the residential microenvironment, which may be considered the most important one, according to the 24-hr mean time spent by urban populations (on average 60–85% of the day) (Klepeis *et al*., 2001; Adgate *et al*., 2002).Various studies have investigated the contribution of indoor and outdoor sources to the observed concentration levels (Huang *et al*., 2007; Kuo *et al*., 2007). The penetration of outdoor-generated particulate pollutants (such as black carbon, BC) into indoor microenvironments has been found to play an important role (Fischer *et al*., 2000; Funasaka *et al*., 2000; Jones *et al*., 2000; Kingham *et al*., 2000; Ho *et al*., 2004; Lazaridis *et al*., 2008). Among the indoor sources present, cooking, smoking and cleaning activities have been found

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to contribute the most (Wallace, 1996; Nazaroff, 2004; Martuzevicius *et al*., 2008; Gaidajis and Angelakoglou, 2009; Lai *et al*., 2010). Smaller particles, mostly in the submicrometer range, are emitted generally through combustion processes, while coarse particles' main source is resuspension caused by people presence and movement (Morawska and Zhang, 2002; He *et al*., 2004). Secondary particles may also be formed in indoor environments through gas-to-particle conversion (Rohr *et al*., 2003).

The majority of the published research deals with PM mass concentrations. Data on residential particle number indoor and outdoor concentrations are relatively scarce (Morawska *et al*., 2003; Matson, 2005; Hussein *et al*., 2006; Wallace, 2006; Diapouli *et al*., 2007; Hoek *et al*., 2008). Moreover, most of the studies have been conducted in unoccupied residences and do not account for indoor sources. They also tend to concentrate on total particle number over specific size ranges rather than examining number size distribution profiles.

The scope of the present work was to characterize PM mass and number concentration levels (PM_{10}, PM_2, ultrafine and accumulation mode particle size distribution, as well as black carbon) at typical residential microenvironments in the centre of Athens. Indoor and outdoor data were statistically analyzed in order to identify the relative contribution of indoor and outdoor sources to the measured indoor concentrations. The effects of specific indoor sources both to particle concentration as well as size distribution were also studied.

MATERIALS AND METHODS

Three typical residences were selected all located in densely populated residential areas in the periphery of Athens city centre. The first and third residences (RES 1 and RES 3) were flats in old multistory buildings in the periphery of the Historical Centre. The second residence (RES 2) was a recently refurbished building, located in a residential area adjacent to a major avenue with high vehicular traffic. Sampling was performed in two campaigns during 2002, covering both cold period (March–April and November–December) and warm period (June–September). Each residence was studied during a period of 1–2 weeks each season.

Custom made gravimetric samplers were employed for the simultaneous measurement of mean 24-hr outdoor PM_{10} and PM_2 concentrations, operating at a flow rate of 24 and 23 L/min, respectively. The PM_{10} head had a round jet impaction stage equivalent to the Reference E.U. Low Volume Sampler (CEN, 1998). The PM_2 sampling head operated as a slit impactor with a cut-off point at 2.1 mm. A two-stage cascade impactor was employed simultaneously to the outdoor samplers, for the measurement of PM_{10} and PM_2 indoor concentrations. PTFE membrane filters were used both indoors and outdoors. All filters were weighed before and after sampling using a microbalance Sartorius BP211D, at controlled conditions of relative humidity and temperature. Outdoor samplers were placed at a height of approximately 8 m above ground level. The indoor sampler was located in the living room of each residence at breathing height (~1.7 m).

Apart from gravimetric samplers, a number of continuous monitors were used as well. Black carbon concentration (BC) was continuously measured by means of an aethalometer (AE-9, Maggee Sci). Moreover, the size resolved aerosol number concentration was obtained by the use of a Scanning Mobility Particle Sizer (SMPS, DMA model 3071 and CPC 3022A, TSI Inc., U.S.A.), for the ultrafine fraction (particles in the size range 10–400 nm), and a Laser Aerosol Spectrometer (Las-x, PMS Inc., U.S.A.) for the accumulation fraction (particles in the size range 100–3000 nm). The SMPS system was operated according to the calibration data sheets supplied by the manufacturer for flow and other operational parameters. Flows within the DMA were checked and adjusted if necessary every 12 h, with the sheath and inlet flows set at 5 and 0.5 L/min respectively. Data reduction and analysis of the recorded size distribution was performed by means of the Aerosol Instrument Manager software (version 4.0, TSI Inc.)

In the case of continuous monitors, sampling of indoor and outdoor air was performed on a 15-minute cycle from a common inlet succeeding an automatic three way-regulating valve, which alternated between two separate indoor and outdoor inlets. Three consecutive 5-min measurements were obtained per cycle. The outdoor inlet extended at least three meters away from the building's external wall and the indoor inlet was located at breathing height in the middle of the living room. In order to minimize particle losses, all tubing was conductive metal of 12 mm internal diameter.

Spot measurements of the ventilation rate were also conducted by means of the SF_6 decay method. One to three measurements for each residence and season were performed. Measurements were conducted under normal living conditions and all daily activities of the residents, as well as relative indoor conditions, were recorded in time-activity diaries. The recorded activities / indoor conditions at each residence and measurement period are summarized in Table 1.

RESULTS AND DISCUSSIONS

Mass Concentration Levels

In the framework of the experimental campaign, PM_{10} and PM_2 mass concentration levels were measured gravimetrically on a 24-hr basis, while black carbon (BC) concentrations were monitored continuously. Descriptive statistics for the 24-hr mean indoor and outdoor concentrations of PM_{10}, PM_2 and BC at each residence and measurement period are presented in Table 2.

The overall PM_{10} 24-hr mean outdoor concentration for all residences was 52.0 ± 14.9 μg/m^3 during the warm period and 53.9 ± 18.4 μg/m^3 during the cold period. Those mean values were above the 24-hr E.U. air quality standard (50 μg/m^3). Outdoor 24-hr mean concentrations at residences #2 and #3 exceeded the limit value of 50 μg/m^3 during approximately half of the measurement days (56% and 43% of the measurement days at RES 2 and RES 3, respectively).

Table 1. Cumulative time for recorded activities/indoor conditions [hrs] at each residence, during warm and cold measurement period.

	Warm period			Cold period			Total
	RES 1	RES 2	RES 3	RES 1	RES 2	RES 3	
Window open	103.0	54.0	25.0	11.0	8.0	30.3	231.3
Air condition on		5.3	2.3		9.0		16.5
Window open/Cooking	1.0	4.3			1.5		6.8
Air condition on/Cooking		1.0			2.5		3.5
Cooking		1.0	1.0	1.8	5.5		9.3
Vacuum cleaning		0.8	0.5				1.3
EMPTY HOUSE	69.3	148.8	46.8	87.8	57.5	50.0	460.0
OCCUPIED HOUSE (no activity)	18.5	23.3	90.0	107.8	79.3	131.5	450.3

Table 2. Descriptive statistics for the PM_{10}, PM_2 and BC 24-hr indoor and outdoor concentration levels [$\mu g/m^3$].

		PM_{10}					
		INDOORS			OUTDOORS		
		Mean	Median	Standard Deviation	Mean	Median	Standard Deviation
Warm period	RES 1	27.1	28.7	6.4	34.8	32.9	6.8
	RES 2	30.8	30.3	8.3	59.1	51.8	27.6
	RES 3	47.3	44.1	10.9	61.9	63.8	10.4
Cold period	RES 1	30.0	23.0	17.0	52.5	44.0	29.7
	RES 2	25.1	23.6	10.1	73.1	70.2	41.3
	RES 3	40.4	40.3	13.8	36.3	31.8	17.6
		PM_2					
Warm period	RES 1	20.7	20.1	4.7	30.2	27.5	7.5
	RES 2	27.3	26.9	6.3	34.7	34.9	13.7
	RES 3	42.4	39.7	8.6	48.7	48.4	8.2
Cold period	RES 1	27.8	23.0	16.6	40.9	37.0	17.5
	RES 2	23.4	23.0	7.5	50.5	39.4	39.5
	RES 3	30.5	29.6	11.8	30.5	28.9	17.1
		BC					
Warm period	RES 1	1.2	1.0	0.7	1.3	1.1	0.7
	RES 2	2.4	2.0	1.3	3.2	2.1	2.1
	RES 3	3.6	3.2	2.1	4.1	3.5	2.8
Cold period	RES 1	1.6	1.3	1.0	2.0	1.6	1.4
	RES 2	2.7	2.1	2.4	3.6	2.5	3.8
	RES 3	3.6	2.4	3.3	3.7	2.6	3.5

Site RES 1 presented a lower percentage of exceedances (25%), which is nevertheless significant considering that the 24-hr air quality standard should not be exceeded for more than 35 days per year (10%). The respective indoor concentration levels were generally lower (mean values for all residences was 35.0 ± 10.7 $\mu g/m^3$ during warm period and 31.8 ± 7.8 $\mu g/m^3$ during cold period), presenting no exceedance of the 50 $\mu g/m^3$ limit value at RES 2 and only a few at RES 1 (5% of the measurement days). Nevertheless, high indoor concentrations, surpassing 50 $\mu g/m^3$, have been recorded at RES 3 during approximately 33% of the measurement days.

Regarding PM_2, the 24-hr mean outdoor value for all three sites was 37.9 ± 9.6 $\mu g/m^3$ during the warm period and 40.6 ± 10.0 $\mu g/m^3$ during the cold period. PM_2 concentrations obtained during this study, are considered close enough to ambient $PM_{2.5}$ (Eleftheriadis *et al*., 2006), in order to use this dataset for an indicative comparison to the annual target value for ambient $PM_{2.5}$ concentrations (25 $\mu g/m^3$), to be achieved by 2015. The measured concentration levels were indeed very high, exceeding this target value during 81%, 71% and 71% at RES 1, RES 2 and RES 3, respectively. Moreover, fine particles contributed significantly to total PM_{10} mass, as shown by the calculated ratio of PM_2-to-PM_{10} concentrations (equal to 0.83, 0.62 and 0.79 at RES 1, RES 2 and RES 3, respectively). High values of the PM_2/PM_{10} concentration ratio are indicative of sites affected by dense vehicular traffic, as was the case of the three studied areas (densely populated residential areas in the periphery of the Athens city centre).

Indoor 24-hr mean PM_2 concentration for all three sites was 30.1 ± 11.1 $\mu g/m^3$ during the warm period and 27.2 ± 3.6 $\mu g/m^3$ during the cold period. In contrast to PM_{10}

indoor levels, the fine particle concentration indoors often exceeded the target value of 25 μg/m^3 (32%, 61% and 80% of the measurement days at RES 1, RES 2 and RES 3, respectively). The high indoor levels may be attributed to the higher penetration ratios of outdoor-generated fine particles indoors and/or to the presence of indoor sources during measurements. Further analysis of the indoor and outdoor concentration time-series, given below, attempts to resolve the relative contribution between indoor and outdoor particle sources.

Outdoors the 24-hr mean BC concentration for all three sites was 2.9 ± 1.4 μg/m^3 during the warm period and 3.1 ± 1.0 μg/m^3 during the cold period. The measured values were typical of urban sites affected by traffic (Hitzenberger and Tohno, 2001; Vianna *et al*., 2007). Indoor concentration levels were somewhat lower than the respective outdoor levels (24-hr mean values equal to 2.4 ± 1.2 μg/m^3 during the warm period and 2.6 ± 1.0 μg/m^3 during the cold period), indicating that outdoor anthropogenic combustion sources are also the primary source of indoor BC (Lunden *et al*., 2008).

Number Concentration Levels

Particle number size distributions were monitored continuously in two size ranges: 10–400 nm and 100–3000 nm. Descriptive statistics for the 24-hr mean indoor and outdoor number concentrations, for each size fraction, are presented in Table 3.

As expected, ambient levels were much lower in the size range 100–3000 nm, since it has been shown that, in polluted environments, particles larger than 100 nm contribute only a small fraction (less than 10%) to the total particle number concentrations (Keywood *et al*., 1999; Shi *et al*., 1999). The highest levels were measured at RES 2, reflecting the effect of heavy traffic (present at the major arterial road located in close vicinity to the measurement site) to the particle number concentration. The measured concentrations were in agreement with results obtained during another measurement campaign in the city of Athens (Diapouli *et al*., 2007). Mean 24-hr number concentrations of particles greater than 10 nm were 2.6 × 10^4 1/cm^3 at a densely populated suburb during cold period and 2.3 × 10^4 1/cm^3 and 4.9 × 10^4 1/cm^3 at a residential area close to the city centre, during warm and cold period respectively. Very high number concentrations of particles larger than 10 nm, similar to the ones recorded at RES 2, have been also reported by Paatero *et al*. (2005) for other Mediterranean urban centers, such as Rome (4.7 × 10^4 1/cm^3) and Barcelona (5.9 × 10^4 1/cm^3).

Indoor 24-hr mean number concentrations were generally lower than the outdoor ones. The highest values were measured at RES 3 instead of RES 2, which displayed the highest outdoor values. Similar results were obtained for PM_{10} and PM_2 indoor concentration levels, indicating increased indoor particle generation at residence # 3.

Indoor - Outdoor Concentrations Relationships

Indoor concentrations from all residences were plotted against the respective outdoor values, for the coarse and fine mass fractions, BC mass concentration (Figs. 1(a)–1(c)) and number concentrations for the fine and accumulation modes of the aerosol size distribution (Figs. 2(a) and 2(b)). The circles on Fig. 2(a) correspond to data points considered as outliers and thus excluded from the regression analysis presented below (Table 4). PM mass concentration data analysis clearly reflected particles' behavior when entering an indoor microenvironment in relation to their size. Indoor and outdoor concentration data for coarse particles ($PM_{2–10}$)

Table 3. Descriptive statistics for the 24-hr indoor and outdoor number concentrations [# 1/cm^3], for particles in the size ranges: 10–400 nm and 100–3000 nm.

		10–400 nm					
		INDOORS (× 10^4 1/cm^3)			OUTDOORS (× 10^4 1/cm^3)		
		Mean	Median	Standard Deviation	Mean	Median	Standard Deviation
Warm period	RES 1	1.4	1.2	0.7	1.9.	1.7	1.0
	RES 2	2.0	1.6	1.3	4.9	2.3	3.4
	RES 3	2.6	1.5	3.4	3.4	3.0	1.9
Cold period	RES 1	2.3	2.0	2.0	2.9	2.6	1.6
	RES 2	1.3	1.2	0.5	5.3	5.1	2.2
	RES 3	3.4	2.1	3.6	4.8	3.4	4.5
		100–3000 nm					
		INDOORS (× 10^3 1/cm^3)			OUTDOORS (× 10^3 1/cm^3)		
		Mean	Median	Standard Deviation	Mean	Median	Standard Deviation
Warm period	RES 1	1.5	1.3	1.0	1.4	1.3	0.5
	RES 2	1.6	1.5	0.7	1.8	1.5	0.8
	RES 3	3.1	2.4	3.2	3.0	2.7	1.3
Cold period	RES 1	1.8	1.6	0.8	2.1	1.8	1.2
	RES 2	2.0	1.5	1.2	2.1	1.4	1.7
	RES 3	1.8	1.3	1.4	2.1	1.6	1.8

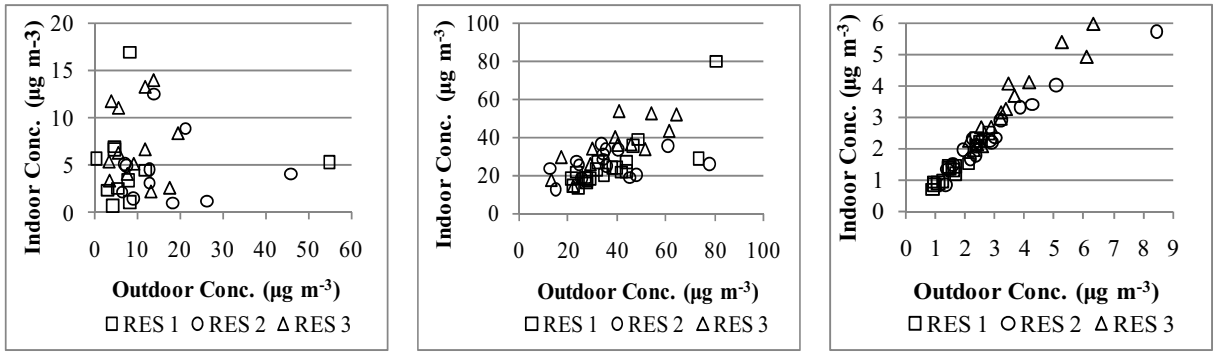

Fig. 1. Regression of indoor and outdoor mass concentration data at all residences, for: (a) $PM_{2–10}$, (b) PM_2 and (c) BC.

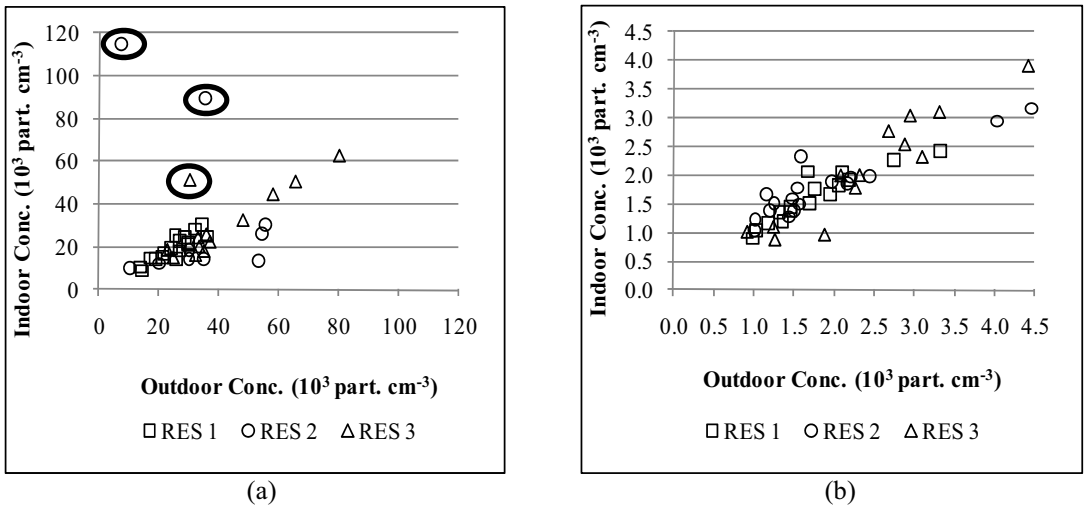

Fig. 2. Regression of indoor and outdoor number concentration data at all residenc.

Table 4. Regression analysis results of indoor versus outdoor concentration data: Correlation coefficient (r), Slope (α), Intercept (β), Ratio of intercept to mean indoor concentration (β/Cin) and Number of data points (N).

	PM_2					BC				
	r	α	β (μg/m³)	β/Cin	N	r	α	β (μg/m³)	β/Cin	N
RES 1	0.81	0.73	–2.3	–0.09	21	0.96	0.89	–0.04	–0.03	16
RES 2	0.30	0.12	21.9	0.86	15	0.98	0.66	0.46	0.19	17
RES 3	0.79	0.60	12.8	0.36	13	0.96	0.90	0.17	0.05	15
All data	0.64	0.51	9.31	0.33	49	0.96	0.81	0.18	0.07	48
	10–400 nm					100–3000 nm				
	r	α	β (×10⁴ 1/cm³)	β/Cin	N	r	α	β (×10³ 1/cm³)	β/Cin	N
RES 1	0.89	0.85	–0.2	–0.08	17	0.92	0.66	0.5	0.28	16
RES 2	0.74	0.32	0.6	0.18	8	0.92	0.54	0.8	0.44	17
RES 3	0.98	0.86	–0.7	–0.24	12	0.73	0.80	0.4	0.15	15
All data	0.84	0.62	0.2	0.06	37	0.81	0.71	0.5	0.24	48

id not exhibit any correlation, suggesting that ambient particle penetration is minimal for this size fraction. The coarse particle concentration indoors may be attributed concentrations, indicating a greater influence of various factors (such as house characteristics, indoor activities and/or meteorological conditions) on the behavior of this size fraction. Regarding particle number concentration data, mostly to indoor particle generation or re-suspension. The indoor/outdoor PM_2 concentrations showed a relatively higher variability at the different sites compared to BC the two size fractions displayed similar patterns, with the exception of some outliers in the case of particles in the size range 10–400 nm, indicative of occasional intense indoor generation for this size fraction.

The results of the regression analysis of indoor and outdoor data for each residence separately, as well as for all three residences, are presented in Table 4 (Coarse particles are excluded, since no correlation was observed for any of the residences.). The linear relationship parameters between indoor and outdoor concentrations were found to correspond to physical factors governing them. In this simplified model the indoor concentration is a function of the outdoor concentration, the latter taken here as an independent variable. More specifically, the correlation coefficient may be used as an indicator of the effect of PM of outdoor origin to the indoor levels. The slope of the regression line corresponds to the fraction of outdoor-generated PM penetrating into the residences, while the intercept (or ratio β/Cin) is a measure of the contribution of indoor sources.

When entering an indoor microenvironment, aerosol particles may be lost on surfaces of cracks and openings due to either settling (coarse particles) or diffusion through Brownian motion (ultrafine particles). Particles in the accumulation mode are expected to penetrate most efficiently since they are least affected by either mechanism (Nazaroff, 2004). This effect was pronounced on the slope values calculated for each size fraction. Ambient aerosol penetration decreased as particle size increased. In the case of number concentration data, slope values calculated for RES 1 and 3 were larger for ultrafines. To the contrary, at RES 2, which was equipped with new technology window frames that enhanced the building's tightness, penetration of ultrafine particles was reduced, due to diffusion losses. Similar slope values calculated for $PM_{2.5}$ and BC (soot) concentration data from the city of Athens have been reported by Hoek *et al*. (2008).

Black carbon presented a distinct behavior from all other aerosol parameters, exhibiting a very good agreement of results between the residences. In particular, BC indoor and outdoor concentration data were highly correlated. Their regression lines presented very high slope values and negligible intercepts, indicative of high particle penetration. A minor deviation was observed in the case of RES 2, where the penetration of outdoor-generated BC was reduced to some extent, while there was also some indoor generation, possibly related to intense oven/grill cooking activities reported for this residence. BC is a chemically stable species mostly found in the accumulation and ultrafine particle modes (Miguel *et al*., 2003). Due to both its size and chemical stability, it is expected to penetrate easily indoors, causing elevated indoor concentration levels (Cao *et al*., 2005; Na and Cocker, 2005).

Ambient particles penetration may be also affected by changes in ventilation rate. During periods of no significant indoor sources particle transfer from outdoors to indoors (expressed as the indoor-to-outdoor concentration ratio, I/O) was examined with respect to the measured ventilation rate. Results for the I/O ratio for the accumulation mode and ultrafine number concentration data, are shown in Fig. 3. The resulting I/O ratios for both size fractions were relatively stable, showing a weak increasing trend, not statistically significant for this data set. Further study is needed in order to assess the relationship governing particles penetration and air exchange rate. Despite the high uncertainty involved in this small set of data points, rendering the observed trend only indicative of a possible relationship, it is interesting to note that ventilation rates in the range of 1.0–1.5 1/h were adequate for the I/O ratio of the accumulation mode aerosol to approach unity for those older residences studied here. Ultrafine particles displayed similar trend but I/O ratios were much lower.

The regression analysis results for each size fraction and residence provided an insight into the measured indoor concentrations. The increased indoor PM mass and number concentrations recorded at RES 3 may be attributed to high penetration of outdoor particles, indicated by large slope value, for all particle categories. Outdoor-generated particles penetration significantly influenced indoor levels at RES 1 as well. The frequent window opening reported by the inhabitants at this residence may have further promoted penetration of the ambient air indoors. At RES 2, in contrast to the other two residences, low slope values were observed, while for PM_2 a poor correlation was found between indoor and outdoor concentration data. The residence construction characteristics (increased building tightness), along with the more frequent mechanical ventilation (use of air conditioning rather than window opening) have resulted in a very low penetration of the ambient PM, for all size fractions.

According to the calculated intercept values, indoor particle generation appears to be the main contributor to the measured indoor concentrations at RES 2 (probably mostly through cooking, whose cumulative time was much larger than for the other two residences). A significant contribution from indoor sources was detected at RES 3 for PM_2 only and may be attributed to particle generation and/or resuspension due to the presence of people for longer time periods. RES 3 was occupied during 66% of the measurement period, while RES 1 and RES 2 were occupied only during 21% and 14% respectively.

Near to real time monitoring of BC and number concentrations allowed for the study of the effect of the recorded indoor activities/conditions to the measured concentration levels. Indoor-to-outdoor concentration ratios were calculated for the specific time periods when an indoor activity or condition was reported. Mean values of these I/O ratios are presented in Fig. 4. The calculated I/O ratios for BC remained rather stable, with a small increase during vacuum cleaning, possibly due to primary graphite carbon emission from the vacuum cleaner motor brushes. The results support our previous assumption that the measured BC was mainly of outdoor origin. Number concentrations of accumulation mode particles also produced stable I/O ratio values, with the exception of cooking, when indoor concentrations exhibited a large increase due to particle generation. The largest I/O variation was observed in the ultrafine particle mode. Both ambient particle penetration indoors and indoor particle generation were affected by the prevailing conditions and activities. Penetration of outdoor-generated particles was

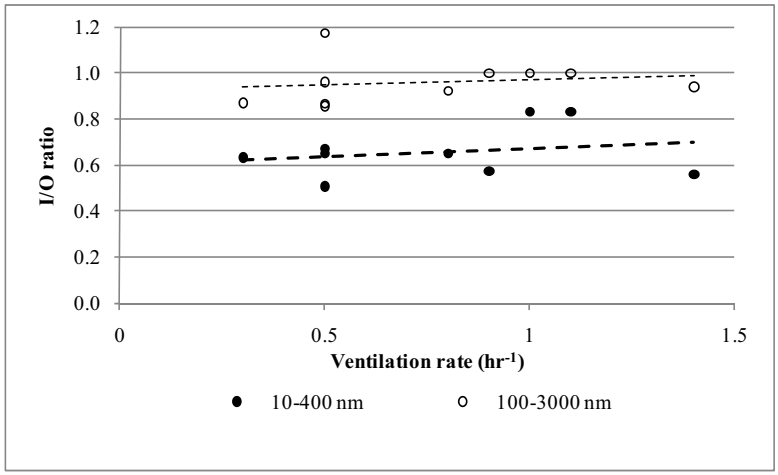

Fig. 3. Indoor-to-outdoor concentration ratio (I/O) as a function of ventilation rate, for ultrafine and accumulation mode particles. Trend lines are only "guides to the eye" and are not statistically significant.

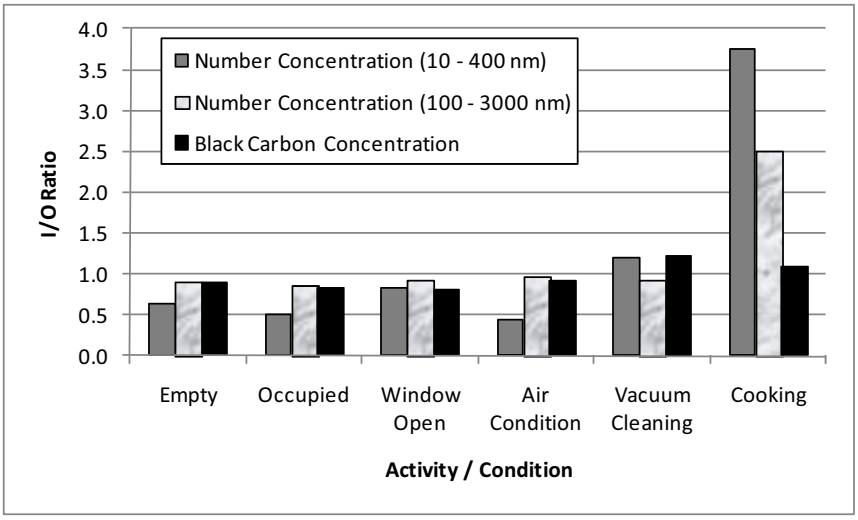

Fig. 4. Indoor-to-outdoor concentration ratio (I/O) as a function of the indoor activities / conditions reported.

enhanced by window opening and decreased by the use of air conditioning. The relatively stable I/O ratios of BC and accumulation mode particles during the different ventilation conditions may be attributed to their efficient penetration indoors. The largest difference in ultrafine particles I/O ratio values corresponded again to cooking and may be attributed to the related combustion processes, which tend to generate particles in the ultrafine and accumulation mode (Long *et al*., 2000; Dennekamp *et al*., 2001; He *et al*. 2004; See and Balasubramanian, 2006).

Apart from indoor concentrations, the relative contribution of indoor and outdoor sources may affect significantly the observed size distribution in the indoor microenvironment. Indoor and outdoor particle number size distributions, normalized with respect to total number concentration (Nt), under different conditions are presented in Figs. 5(a)–5(d). In the first two cases, outdoor total number concentration was larger than the respective indoor one, indicating the absence of significant indoor sources. The indoor and outdoor particle size distributions exhibited, nevertheless, very different patterns. There was a shift towards larger particle diameters when outdoor-generated particles were infiltrating indoors, more pronounced when the residence was occupied. This phenomenon may be mostly attributed to infiltration and other losses for the lower sizes of the ultrafine fraction. In the case of the vacuum cleaning event (Fig. 5(c)), indoor and outdoor concentrations were similar, due to window opening during vacuuming. Cooking, as expected, caused increased indoor number concentrations, again shifting the size distribution towards larger particle diameters. The effect of cooking may be seen more clearly in Fig. 6, where the dynamic evolution of aerosol number distribution after a cooking event is presented. The freshly emitted during cooking ultrafine particles are seen to undergo rapid coagulation, resulting in a transfer of particle mass from the ultrafine to the accumulation mode, while total number concentration decreases (Nazaroff, 2004). This kind of particle size distribution data may provide a much clearer picture of the transport, losses and

generation of particles of different size fractions. In addition they may be used for inhalation dosimetry calculations during specific indoor source events (Mitsakou *et al*., 2007).

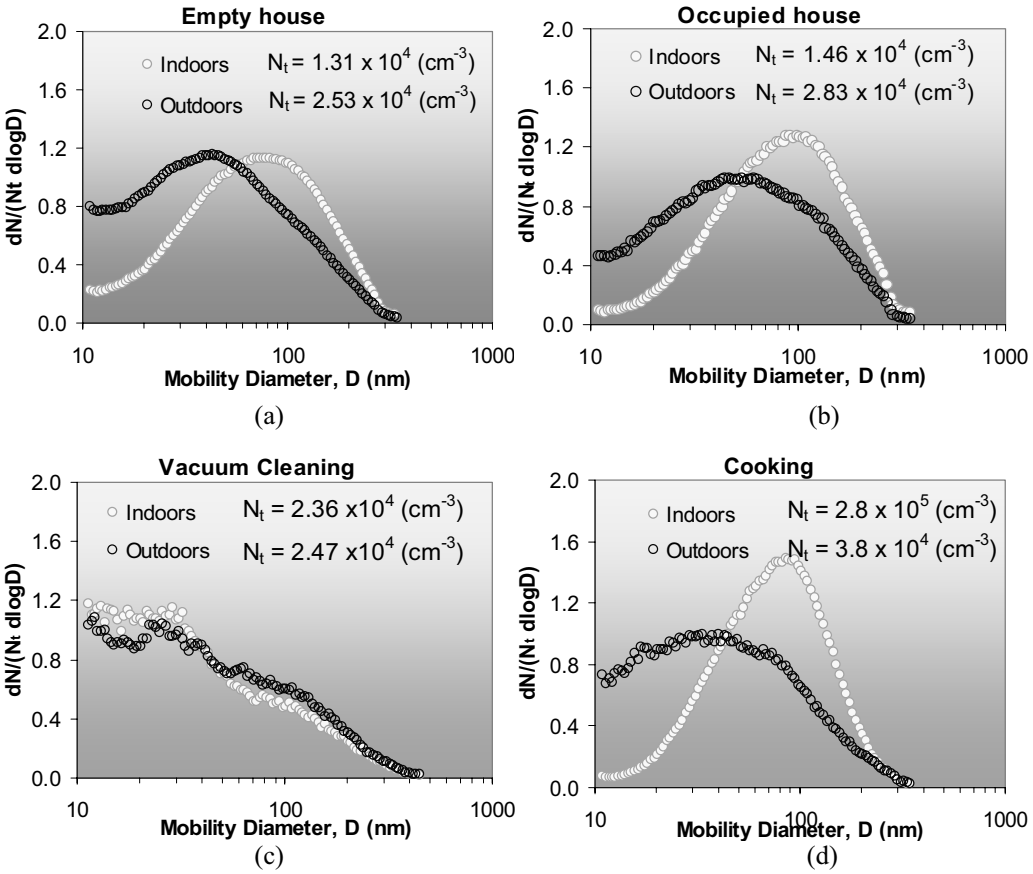

Fig. 5. Size distribution of indoor and outdoor particles, when the residence is empty (a) and occupied (b) and during vacuuming cleaning (c) and cooking (d).

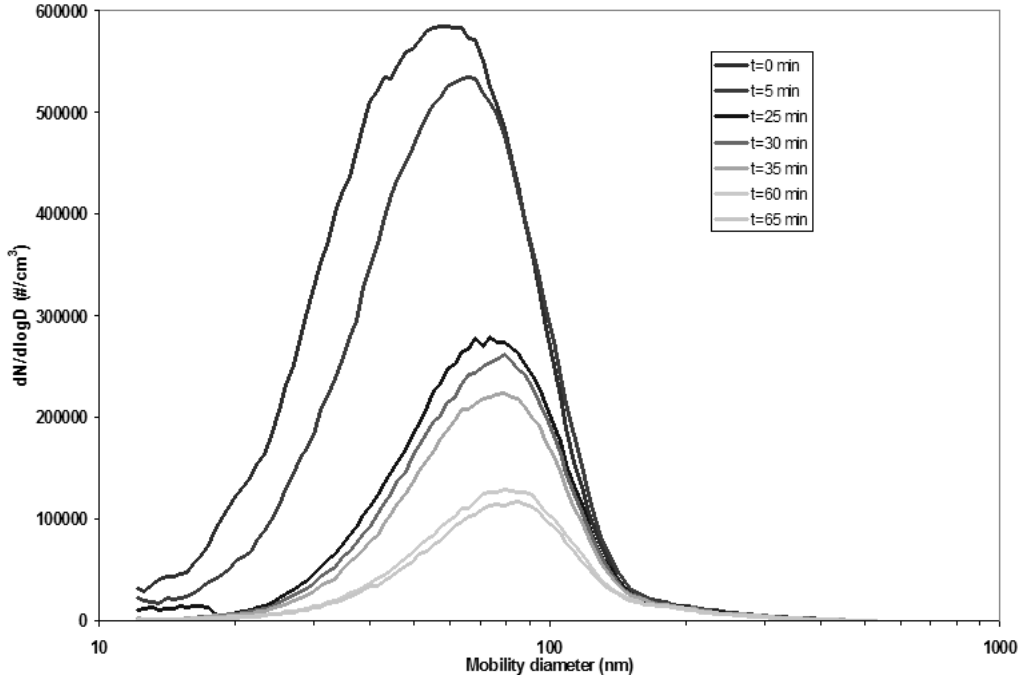

Fig. 6. Detailed evolution of the number size distribution during a cooking event.

CONCLUSION

The scope of this work was to examine particle concentration of different size fractions and composition (PM_{10}, PM_2, ultrafine and accumulation mode particles, as well as black carbon) at typical residential microenvironments and to account for the specific factors that control their concentration levels and size distribution.

The measured outdoor levels at the three residential flats under study, all located in densely populated residential areas in the periphery of the Athens city centre, were found significant with respect to the relevant PM limit values. Mean 24-hr PM_{10} concentrations exceeded the limit value of 50 $\mu g/m^3$ during approximately half of the measurement days at two of the residences. The respective PM_2 concentrations were above the annual target value of 25 $\mu g/m^3$ during 70–80% of the days, at all residences. Outdoor BC and particle number concentration levels were also high, typical of urban sites affected by heavy traffic.

Indoor levels were generally lower than the corresponding outdoor ones. Nevertheless, elevated indoor concentrations were recorded, caused by increased ambient aerosol penetration in the indoor microenvironments and/or indoor particle generation. Regression analysis of indoor and outdoor data revealed that BC measured indoors, may be mainly considered of outdoor origin. This specific chemical species exhibits high penetration efficiency, due to its size and chemical stability. A large fraction of the outdoor-generated PM_2, ultrafine and accumulation mode particles also penetrated effectively indoors, causing elevated indoor levels. In contrast, coarse particles indoor concentration was mainly attributed to indoor sources (particle generation or re-suspension). Particle generation by routine indoor activities contributed to PM_2 and ultrafine particles indoor concentrations. Cooking appeared the strongest contributor, significantly affecting indoor levels of all particle size fractions.

Residential microenvironments play a significant role in population total personal exposure. The identification of the possible indoor sources present, as well as the influence of the ambient particulate pollution, is crucial in order to develop effective control measures. Building structure (such as materials, ventilation systems), as well as carefully selected housekeeping practices, may greatly contribute towards an integrated policy for the protection of public health, especially in big urban centers where particulate pollution remains a major environmental issue.

REFERENCES

Adgate, J.L., Ramachandran, G., Pratt, G.C., Waller, L.A. and Sexton, K. (2002). Spatial and Temporal Variability in Outdoor, Indoor, and Personal $PM_{2.5}$ Exposure. *Atmos. Environ.* 36: 3255–3265.

Brown, L. (1983). National Radiation Survey in the UK: Indoor Occupancy Factors. *Radiat. Prot. Dosim.* 5: 203–208.

Cao, J.J., Lee, S.C., Chow, J.C., Cheng, Y., Ho, K.F., Fung, K., Liu, S.X. and Watson, J.G. (2005). Indoor/Outdoor Relationships for $PM_{2.5}$ and Associated Carbonaceous Pollutants at Residential Homes in Hong Kong - Case Study. *Indoor Air* 15: 197–204.

CEN, European Committee for Standardization. (1998). Air Quality - Determination of the PM_{10} Fraction of Suspended Particulate Matter. Reference Method and Field Test Procedure to Demonstrate Equivalence of Measurement Methods, EN 12341.

Dennekamp, M., Howarth, S., Dick, C.A.J., Cherrie, J.W., Donaldson, K. and Seaton, A. (2001). Ultrafine Particles and Nitrogen Oxides Generated by Gas and Electric Cooking. *Occup. Environ. Med.* 58: 511–516.

Diapouli, E., Chaloulakou, A. and Spyrellis, N. (2007). Concentration Levels of Ultrafine Particles in different Microenvironments – Implications to Children Exposure. *Sci. Total Environ.* 388: 128–136.

Donaldson, K., Brown, D., Clouter, A., Duffin, R., Macnee, W., Renwick, L., Tran, L. and Stone, V. (2002). The Pulmonary Toxicology of Ultrafine Particles. *J. Aerosol Med.* 15: 213–220.

Eleftheriadis, K., Colbeck, I., Housiadas, C., Lazaridis, M., Mihalopoulos, N., Mitsakou, C., Smolík, J. and Ždímal, V. (2006). Size Distribution, Composition and Origin of the Submicron Aerosol in the Marine Boundary Layer during the Eastern Mediterranean "SUB-AERO" Experiment. *Atmos. Environ.* 40: 6245–6260.

Fischer, P., Hoek, G., Van Reeuwijk, H., Briggs, D., Lebret, E., Van Wijnen, J., Kingham, S. and Elliott, P. (2000). Traffic-Related Differences in Outdoor and Indoor Concentrations of Particles and Volatile Organic Compounds in Amsterdam. *Atmos. Environ.* 34: 3713–3722.

Funasaka, K., Miyazaki, T., Tsuruho, K., Tamaru, K., Mizuno, T. and Kuroda, K. (2000). Relationship between Indoor and Outdoor Carbonaceous Particulates in Roadside Households. *Environ. Pollut.* 110: 127–134.

Gaidajis, G. and Angelakoglou, K. (2009). Indoor Air Quality in University Classrooms and Relative Environment in Terms of Mass Concentrations of Particulate Matter. *J. Environ. Sci. Health., Part A* 44: 1227–1232.

He, C., Morawska, L., Hitchins, J. and Gilbert, D. (2004). Contribution from Indoor Sources to Particle Number and Mass Concentrations in Residential Houses. *Atmos. Environ.* 38: 3405–3415.

Hitzenberger, R. and Tohno, S. (2001). Comparison of Black Carbon (BC) Aerosols in Two Urban Areas - Concentrations and Size Distributions. *Atmos. Environ.* 35: 2153–2167.

Ho, K.F., Cao, J.J., Harrison, R.M., Lee, S.C. and Bau, K.K. (2004). Indoor/Outdoor Relationships of Organic Carbon (OC) and Elemental Carbon (EC) in $PM_{2.5}$ in Roadside Environment of Hong Kong. *Atmos. Environ.* 38: 6327–6335.

Hoek, G., Kos, G., Harrison, R., de Hartog, J., Meliefste, K., ten Brink, H., Katsouyanni, K., Karakatsani, A., Lianou, M., Kotronarou. A., Kavouras, I., Pekkanen, J., Vallius, M., Kulmala, M., Puustinen, A., Thomas, S., Meddings, C., Ayres, J., van Wijnen, J. and Hameri, K.

(2008). Indoor–Outdoor Relationships of Particle Number and Mass in four European Cities. *Atmos. Environ.* 42: 156–169.

Huang, H., Cao, J.J., Lee, S.C., Zou, C.W., Chen, X.G. and Fan, S.J. (2007). Spatial Variation and Relationship of Indoor/Outdoor $PM_{2.5}$ at Residential Homes in Guangzhou City, China. *Aerosol Air Qual. Res.* 7: 518–530.

Hussein, T., Glytsos, T., Ondracek, J., Dohanyosova, P., Zdimal, V., Hameri, K., Lazaridis, M., Smolik, J. and Kulmala, M. (2006). Particle Size Characterization and Emission Rates during Indoor Activities in a House. *Atmos. Environ.* 40: 4285–4307.

Ibald-Mulli, A., Wichmann, H.E., Kreyling, W. and Peters, A. (2002). Epidemiological Evidence of Health Effects on Ultrafine Particles. *J. Aerosol Med.* 15: 189–201.

Jones, N.C., Thornton, C.A., Mark, D. and Harrison, R.M. (2000). Indoor/Outdoor Relationships of Particulate Matter in Domestic Homes with Roadside, Urban and Rural Locations. Atmos. Environ. *Atmos. Environ.* 34: 2603–2612.

Keywood, M.D., Ayers, G.P., Gras, J.L., Gillett, R.W. and Cohen, D.D. (1999). Relationships between Size Segregated Mass Concentration Data and Ultrafine Particle Number Concentrations in Urban Areas. *Atmos. Environ.* 33: 2907–2913.

Kingham, S., Briggs, D., Elliott, P., Fischer, P. and Lebret, E. (2000). Spatial Variations in the Concentrations of Traffic-Related Pollutants in Indoor and Outdoor Air in Huddersfield, England. *Atmos. Environ.* 34: 905–916.

Klepeis, N.E., Nelson,W.C., Ott,W.R., Robinson, J.P., Tsang, A.M., Switzer, P., Behar, J.V., Hern, S.C. and Engelmann, W.H. (2001). The National Human Activity Pattern Survey (NHAPS): A Resource for Assessing Exposure to Environmental Pollutants. *J. Exposure Anal. Environ. Epidemiol.* 11: 231–252.

Kuo, Y.-M., Hung, H.F. and Yang, T.T. (2007). Chemical Compositions of $PM_{2.5}$ in Residential Homes of Southern Taiwan. *Aerosol Air Qual. Res.* 7: 403–416.

Lai, S.C., Ho, K.F., Zhang, Y.Y., Lee, S.C., Huang, Y. and Zou, S.C. (2010). Characteristics of Residential Indoor Carbonaceous Aerosols: A Case Study in Guangzhou, Pearl River Delta Region. *Aerosol Air Qual. Res.* 10: 472–478.

Lazaridis, M., Aleksandropoulou, V., Hanssen, J.E., Dye, C., Eleftheriadis, K. and Katsivela, E. (2008). Inorganic and Carbonaceous Components in Indoor/Outdoor Particulate Matter in two Residential Houses in Oslo, Norway. *J. Air Waste Manage. Assoc.* 58: 346–356.

Long, C.M., Suh, H.H and Koutrakis, P. (2000). Characterization of Indoor Particle Sources using Continuous Mass and Size Monitors. *J. Air Waste Manage. Assoc.* 50: 1236–1250.

Lunden, M.M., Kirchstetter, T.W., Thatcher, T.L., Hering, S.V. and Brown, N.J. (2008). Factors Affecting the Indoor Concentrations of Carbonaceous Aerosols of Outdoor Origin. *Atmos. Environ.* 42: 5660–5671.

Martuzevicius, D, Grinshpun, S.A., Lee, T., Hu, S., Biswas, P., Reponen, T. and LeMasters, G. (2008). Traffic-Related $PM_{2.5}$ Aerosol in Residential Houses Located near Major Highways: Indoor versus Outdoor Concentrations. *Atmos. Environ.* 42: 6575–6585.

Matson, U. (2005). Indoor and Outdoor Concentrations of Ultrafine Particles in some Scandinavian Rural and Urban Areas. *Sci. Total Environ.* 343: 169–176.

Miguel, A.H., Eiguren-Fernandez, A., Jaques, P.A., Froines, J.R., Grant, B.L., Mayo, P.R. and Sioutas, C. (2003). Seasonal Variation of the Particle Size Distribution of Polycyclic Aromatic Hydrocarbons and of Major Aerosol Species in Claremont, California. *Atmos. Environ.* 38: 3241–3251.

Mitsakou, C., Housiadas, C., Eleftheriadis, K., Vratolis, S., Helmis, C. and Asimakopoulos, D. (2007). Lung Deposition of Fine and Ultrafine Particles Outdoors and Indoors during a Cooking Event and a No Activity Period. *Indoor Air* 17: 143–152.

Morawska, L. and Zhang, J. (2002). Combustion Sources of Particles: Health Relevance and Source Signatures. *Chemosphere* 49: 1045–1058.

Morawska, L., He, C., Hitchins, J., Mengersen, K. and Gilbert, D. (2003). Characteristics of Particle Number and Mass Concentrations in Residential Houses in Brisbane, Australia. *Atmos. Environ.* 37: 4195–4203.

Na, K. and Cocker III, D.R. (2005). Organic and Elemental Carbon Concentrations in Fine Particulate Matter in Residences, Schoolrooms, and Outdoor Air in Mira Loma, California. *Atmos. Environ.* 39: 3325–3333.

Nazaroff, W.W. (2004). Indoor Particle Dynamics. *Indoor Air* 14 (Suppl 7): 175–183.

Paatero, P., Aalto, P., Picciotto, S., Bellander, T., Castano, G., Cattani, G., Cyrys, J., Kulmala, M., Lanki, T., Nyberg, F., Pekkanen, J., Peters, A., Sunyer, J., Forastiere, F. and The HEAPSS Study Group. (2005). Estimating Time-Series of Aerosol Particle Number Concentrations in the Five HEAPSS Cities on the Basis of Measured Air Pollution and Meteorological Variables. *Atmos. Environ.* 39: 2261–2273.

Pope, C.A. (2000). Review: Epidemiological Basis for Particulate Air Pollution Health Standards. *Aerosol Sci. Technol.* 32: 4–14.

Pope, C.A. and Dockery, D.W. (2006). Health Effects of Fine Particulate Air Pollution: Lines that Connect. *J. Air Waste Manage. Assoc.* 56: 709–742.

Pope, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K. and Thurston, G.D. (2002). Lung Cancer, Cardiopulmonary Mortality, and Long-term Exposure to Fine Particulate Air Pollution. *J. Am. Med. Assoc.* 287: 1132–1141.

Rohr, A.C., Weschler, C.J., Koutrakis, P. and Spengler, J.D. (2003). Generation and Quantification of Ultrafine Particles through Terpene/Ozone Reaction in a Chamber Setting. *Aerosol Sci. Technol.* 37: 65–78.

See, S.W. and Balasubramanian, R. (2006). Physical Characteristics of Ultrafine Particles Emitted from Different Gas Cooking Methods. *Aerosol Air Qual. Res.* 6: 82–92.

Shi, J.P., Khan, A.A. and Harrison, R.M. (1999). Measurements of Ultrafine Particle Concentration and

Size Distribution in the Urban Atmosphere. *Sci. Total Environ.* 235: 51–64.

Vianna, M., Maenhaut, W., ten Brink, H.M., Chi, X., Weijers, E., Querol, X., Alastuey, A., Mikuska, P. and Vecera, Z. (2007). Comparative Analysis of Organic and Elemental Carbon Concentrations in Carbonaceous Aerosols in three European Cities. *Atmos. Environ.* 41: 5972–5983.

Wallace, L. (1996). Indoor Particles: A Review. *J. Air Waste Manage. Assoc.* 46: 98–126.

Wallace, L. (2006). Indoor Sources of Ultrafine and Accumulation Mode Particles: Size Distributions, Size-Resolved Concentrations, and Source Strengths. *Aerosol Sci. Technol.* 40: 348–60.

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