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Number Concentrations and Modal Structure of Indoor/Outdoor Fine Particles in Four European Cities

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ABSTRACT

Indoor/outdoor aerosol size distribution was measured in four European cities (Oslo–Norway, Prague–Czech Republic, Milan–Italy and Athens–Greece) during 2002 in order to examine the differences in the characteristics of the indoor/outdoor modal structure and to evaluate the effect of indoor sources to the aerosol size distributions. All the measurement sites were naturally ventilated and were occupied during the campaigns by permanent residents or for certain time periods by the technical staff responsible for the instrumentation. Outdoor particle number (PN) concentrations presented the higher values in Milan and Athens (median values 1.4×10^4 # cm^{-3} and 2.9×10^4 # cm^{-3} respectively) as a result of elevated outdoor emissions and led to correspondingly higher indoor values compared to Oslo and Prague. In absence of indoor activities, the indoor concentrations followed the fluctuations of the outdoor concentrations in all the measurement sites. Indoor activities (cooking, smoking, etc.) resulted in elevated indoor PN concentrations (maximum values ranging between 1.7×10^5 # cm^{-3} and 3.2×10^5 # cm^{-3}) and to I/O ratios higher than one. The I/O ratios were size dependant and for periods without indoor activities, they presented the lowest values for particles < 50 nm (0.51 ± 0.15) and the ratios increased with fine particle size (0.79 ± 0.12 for particles between 100–200 nm). The analysis of the modal structure showed that the indoor aerosol size distribution characteristics differ from the outdoors under the effect of indoor sources. The percentage of unimodal size distributions increased during indoor emissions, compared to periods without indoor sources, along with the number concentration of Aitken mode particles, indicating emissions in specific size ranges according to the type of the indoor source.

Keywords: Indoor/Outdoor aerosol; I/O ratio; Modal structure; Indoor sources.

INTRODUCTION

A number of epidemiological studies revealed various correlations between high particulate matter (PM) concentrations and adverse health effects, including respiratory symptoms, asthma and allergies, decreased lung function, cardiovascular disorders and increased mortality (Pope *et al.*, 2009; Bentayeb *et al.*, 2012; Kim *et al.*, 2015; Maynard, 2015; Wang *et al.*, 2016). In the last two decades, many studies showed that exposure to aerosol occurs both indoors and outdoors since people nowadays, especially in big cities, spend most of their time in indoor environments (Eurostat, 2004; Kleipis *et al.*, 2001). Indoor aerosols originate both from the outdoor environment and from indoor sources (Jones 1999), with the effect of the latter being particularly important and resulting in PM concentrations higher than outdoors (Hussein *et al.*, 2005; Wallace *et al.*, 2006). The health effects of indoor aerosols are not yet well characterized and their consequences on humans depend on many factors such as type of indoor environment, chemical composition

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of aerosol, duration of exposure and physical characteristics of the exposed individuals (Lai *et al.*, 2004; Cheng *et al.*, 2012; Beko *et al.*, 2015; Slezakova *et al.*, 2015; Spilak *et al.*, 2015; Sunyer *et al.*, 2015).

In most indoor air quality studies, the PM levels are expressed by their indoor mass concentrations in different size fractions (PM_{10}, $PM_{2.5}$, or PM_1). Many of these studies were performed in indoor and outdoor environments of residential areas (Hänninen *et al.*, 2004; Baxter *et al.*, 2007; Kuo *et al.*, 2007; Rodes *et al.*, 2010). However, mass concentration measurements are not representative of the presence of smaller particles (especially for ultrafine particles - UFP) that contribute very little to particle mass but they are dominant when particle number (PN) concentrations are considered. UFP are produced in indoor environments due to a vast range of common everyday activities such as cooking, cleaning, smoking (even electronic cigarettes), burning of candles and incense sticks, etc., (Buonano *et al.*, 2009; Gao *et al.*, 2013; Wu *et al.*, 2011; Zhao *et al.*, 2016). Indoor sources of UFP also include emissions from electric appliances, office equipment and printers, washing powders and household cleaning products (Destaillats *et al.*, 2008; Wensing et *al.*, 2008; Shcipp *et al.*, 2011, 2012). The particle's penetration efficiency into the human respiratory tract is size dependant and has its maxima in the size range of 100 nm - 1 μm (Housiadas and Lazaridis 2010; Lazaridis *et al.*, 2001; Hussein *et al.*, 2015) and therefore these particles can be more dangerous to human health (Oberdörster, 2001; Franck *et al.*, 2011; Heinzerling *et al.*, 2016). These findings prompted many researchers, in recent years, to focus on PN concentrations in residential areas (Bhangar *et al.*, 2011; Kerney *et al.*, 2011; Mullen *et al.*, 2011) and, furthermore, to estimate the contribution of the outdoor environment and of the indoor sources to the indoor particle load (Morawska *et al.*, 2001; He *et al.*, 2004; Matson, 2005; Beko *et al.*, 2013; Talbot *et al.*, 2016; Tong *et al.*, 2016). Nevertheless, few studies have presented a complete analysis of the modal structure of indoor aerosols and highlighted the differences between indoor and outdoor size distributions characteristics in geographically different large cities within Europe (Hussein *et al.*, 2005; Zhu *et al.*, 2005).

This study presents PN concentration measurements in four European cities (Oslo-Norway, Prague-Czech Republic, Milan-Italy and Athens-Greece). The collected data were used for the analysis of the indoor and outdoor size distributions. The focus of this work is on the characteristics of the indoor to outdoor number size distributions and on estimating the effect of indoor sources to indoor aerosol modal structure.

MATERIALS AND METHODS

Measurement Sites

The indoor/outdoor PN concentration measurements were performed at the metropolitan areas of Athens, Milan, Prague and Oslo as part of work performed within the framework of the multi-centre Characterization of Urban Air Quality Indoor/Outdoor Particulate Matter Chemical Characteristics and Source-to-Inhaled Dose Relationships (URBAN – AEROSOL) study. The measurement sites were selected to study and compare indoor to outdoor correlations of PM and gaseous pollutants in different geographical areas in Europe. In Athens and Milan the sites were located at or close to the city center and were characterized by high emissions, mainly by traffic related combustion aerosols. The Oslo and Prague sites were located at suburban residential areas. Outdoor particle number concentrations are not directly comparable between cities since they are strongly influenced by the specific characteristics of each site (vehicular load, nearby emissions, height, meteorological conditions, etc.). Outdoor concentrations were mainly used to examine the changes of outdoor aerosol characteristics after entering to the indoor environment and to evaluate the effect of indoor sources to the aerosol size distributions. Air exchange rate measurements were conducted in Oslo, Milan and Athens using SF_6 as a tracer gas. More specifically, SF_6 gas was released at a constant rate from a portable release system and air samples were taken with syringes over short time intervals (10 min) and analysed by a portable gas chromatograph. The air exchange rate was determined by observing the concentration decay vs. time of the tracer gas released into the room. The particle number size distribution measurements were conducted in all the sites with the same Scanning Mobility Particle Sizer (SMPS 3934C, TSI Inc., USA). Measurement size ranges during each campaign corresponded to operating parameters set by measurement staff (flows, timing, inlet impactor, etc.) and to specific needs of each campaign. More specifically, the measured size ranges were 10–470 nm (Oslo), 14–552 nm (Prague) and 14–764 nm (Milan). In Athens the measurements were conducted in three different periods and the size ranges of the SMPS were 12–533 nm during March–April 2002, 11–461 nm during August–September 2002 and 14–737 nm during November–December 2002. The SMPS system worked with a 180 s upward scan, followed by 60 s downward scan and another 60 s interval for flushing of the sampling train. The sample flow rates into the SMPS and the sheath flow rates varied between 0.3–0.6 L min^{-1} and 3–6 L min^{-1} respectively, according to the specific needs of each campaign. Size dependant particle losses in the tubing were calculated for the measurements in Prague, Oslo and Milan according to the specific characteristics of the sampling lines (diameter, number of bends, flow rate, etc.) and the data were corrected accordingly. In Athens particle losses were minimized by using conductive metal tubing of 12 mm diameter. Particle losses due to diffusion in the DMA in all sites were incorporated by selecting the correction option in the TSI Aerosol Instrument Manager (AIM) software, which is standard operation procedure in quality assurance of size distribution measurements.

The same instrument was used for sampling both indoors and outdoors using a valve system that allowed the aerosol samples to be drawn in alternating 15 or 20 min periods (3 consecutive 5 min periods in Oslo and Athens and 4 consecutive 5 min periods in Prague and Milan) from indoors and outdoors. The measurement time periods were selected in order to obtain at least 3 consecutive and directly comparable size distributions indoors and outdoors, which resulted in better interpretation of the collected data and

quicker detection of possible instrument malfunctions. The valve operated so that it alternated between the indoor and outdoor inlets in 5 seconds without creating a pressure drop (Teflon rotating ball valve) in the sampling lines. The supply of the indoor or outdoor aerosol during measurements was therefore undisturbed and free from obstructions and losses. Although the use of one SMPS system for both locations (indoor and outdoor) has the disadvantage of not providing simultaneous measurements of the indoor and the outdoor environment, it offers more reliable data for comparison since it is practically impossible to find two different instruments that measure the same PN concentrations in real life conditions. Furthermore, detailed intercomparisons of the two instruments should be necessary before each campaign, both in controlled conditions and in the actual measurement sites which would significantly increase the duration and cost of the campaigns.

Oslo: Suburban-Residential

The measurements were carried out on the ground floor (approximately 41 m^2) in a two storey wooden house in the suburbs of Oslo (Lazaridis *et al.*, 2008) during the summer period (June 3–June 17, 2002). The house was furnished and naturally ventilated. The measurements were conducted in the first floor, which was isolated, during the campaign, from the second floor via two permanently closed doors. The kitchen, sitting room and cellar were located at the ground floor and the instrumentation was placed in the sitting room. The door leading to the cellar was also permanently closed. There were 3 windows in the ground floor. The house was unoccupied by permanent residents but the measurement staff remained in the house from about 09:00 until noon for the purpose of measurement follow up and maintenance. These time periods were also characterised by frequently opening and closing of windows and doors. The total daily time that windows were open ranged from 90 min to 150 min and was limited between 09:00 and 14:00. Windows were closed at the time periods that the indoors sources were active, and remained closed at least two hours after the end of the activities. The air exchange rate varied between 0.79 h^{-1} and 1.97 h^{-1} with average value 1.15 ± 0.34 h^{-1}. During the measurement campaign the outdoor temperature and relative humidity ranged from 9.7 to 26.2°C and 28 to 94% with average values 15.7°C and 57%, respectively. The average wind speed during the measurement's period was 1 m s^{-1}, and varied between 0.1 and 2 m s^{-1}.

Prague: Suburban-Residential

The particle size distribution measurements in Prague were performed during November 16th–29th 2002 (Smolík *et al.*, 2008) in a naturally ventilated guest house of the Institute of Chemical Process Fundamentals (Academy of Sciences of the Czech Republic) at the suburbs of Prague. The area has only minor traffic influence (Ondráček *et al.*, 2011). The apartment was on the first floor of a two floor house and it was occupied during the campaign by a couple. The two floors of the building were separate brick constructions and did not have a common entrance. The apartment consisted of hallway corridor, two bedrooms, a living room and a kitchen. The instrumentation was placed in the living room, which had one wooden frame window. During the measurements, the central heating was always on, which resulted in indoor temperatures between 23.9°C and 27.4°C. A detailed log of the couple's activities was kept during the measurement period. Windows were closed during the measurement campaign except for a time period of 45 minutes on the last day of the campaign. According to the basic meteorological parameters (ambient temperature, and relative humidity), the weather conditions were calm most of the time (the average wind speed was 0.5 m s^{-1}, varying between 0–2 m s^{-1}) the average outdoor temperatures was 7.5°C (varying between 4.3–11.1°C) and the average outdoor relative humidity was 85.9% (varying between 69.7–93.1%).

Milan: Urban-Traffic

The Milan measurement spot was located in an office on the ninth floor of a building in the centre of Milan. The floor consisted of ten offices, one big conference room and four bathrooms. The measurements were carried out during the autumn period (October 11–October 21, 2002). The office was used as a measurement site and the presence of people was limited to the measurement staff, mostly in the morning during working hours. There were two metal frame windows in the office, which were open for 1–2 hours every day, except of the last day of the campaign, when they remained open for the whole day. The instrumentation was placed in the nearby office and the sampling lines were placed inside a special designed metal construction which connected the two offices via the outdoor environment. The tubing inputs to the measurements office were carefully insulated. Three air exchange rates measurements were conducted and the calculated values varied between 0.54 h^{-1} and 2.03 h^{-1}. The value 2.03 was obtained on a very windy day. The average outdoor relative humidity was 81.4% (varying between 23.3–100%), the average outdoor temperature was 15.3°C (varying between 8.8–21.1°C) and (the average wind speed was 1.3 m s^{-1} (varying between 0.4–3.5 m s^{-1}).

Athens: Urban-Traffic

Three typical residences (three flats each at the 4th floor of three different blocks of flats) were selected for measurements, all located in densely populated residential areas at the periphery of the city centre. All residences were influenced by vehicular traffic. The residences were constructed between 1950 and 1985 and their total area varied between 28 m^2 and 80 m^2. Further details of the measurement sites in Athens can be found in Halios *et al.* (2009). Sampling was performed during two campaigns, between March and December 2002, covering both warm and cold season and each residence was studied during a period of 1–2 weeks for two seasons of the year. Since the residences were inhabited (1 person in each residence), the time periods that windows were open varied in each apartment according to the specific needs of the inhabitants and to the season of the year. The exact time periods that

windows were open were register in a detailed activities log book. The percentage of the time that windows were open relative to the total measurements' time, ranged from 2% to 29%. Air exchange rates varied between 0.3 h^{-1} and 0.5 h^{-1} in the winter period and between 0.8 h^{-1} and 1.4 h^{-1} in the summer period. During the measurements, meteorological parameters were monitored by a small mast 5 meters high on the roof of the buildings. Outdoor average temperature and relative humidity were 22.7 ± 2.8°C and 55 ± 17% in the winter periods with the wind speed at 1.8 ± 1.4 m s^{-1} The corresponding average and standard deviation values in the summer periods were 12.9 ± 2.6°C, 70 ± 12% and 1.7 ± 1.8 m s^{-1} respectively.

Indoor Activities in the Different Measurement Sites

The apartment in Oslo was uninhabited and in Milan an office was selected as a measurements site. In order to examine the effect of indoor sources to particle concentrations, several simulated activities (Table 1) were performed. In Oslo, the activities were conducted during 8 out of the 15 measurements days, between 11:00 and 13:00. In three cases, two activities took place simultaneously (Frying and smoking, frying and vacuum cleaning, frying and smoking). The effect of each activity to the indoor particles concentrations was not examined separately and the total monitoring time was divided into time periods with indoor activities and time periods without indoor activities. The same approach was used in all the measurements sites. In Milan limited simulated indoor activities were performed (Table 1), so the indoor particles characteristics were mostly influenced by the presence of people and by the infiltration of outdoor particles. On the other hand, the residences in Athens and Prague were inhabited and the measurements corresponded to real life conditions. All the indoor activities were registered in detailed log books and they are presented in Table 1. Many of the activities were conducted simultaneously and the repeatability of each activity regarding its specific characteristics was obviously very limited.

Data Processing

The raw data of the particle number size distributions were interpolated to a common time using cubic spline interpolation. All computations were performed in Matlab 7.10 (R2010). Furthermore, the data set were thoroughly examined in order to exclude values originating from instruments malfunctions. The evaluation of the particle number size distributions has been performed with specially designed aerosol algorithms for identifying and calculating the variables that describe the multi log-normal size distributions (Ždímal *et al.*, 2008; Ondráček *et al.*, 2009). Each mode in a multi log-normal size distribution is characterized by the geometric mean diameter (GMD), the geometric standard deviation (GSD) and the PN in this mode (Hinds, 1999). The multi log-normal distribution function has been commonly used to parameterize the particle number size distributions indoors and outdoors (Birmili *et al.*, 2001; Hussein *et al.*, 2005).

RESULTS AND DISCUSSION

An Overview of Outdoor Particle Number Concentrations

Although the duration of the measurement campaigns (two weeks in each site) is short and might not be sufficient to be representative of the pollution characteristics of each city in general, the results give an impression about the day-to-

Table 1. Frequency of indoor activities that were conducted in the different sites during the measurements campaigns and percentages of time with indoor activities relative to total monitoring time.

Indoor Activity	Frequency of indoor activities			
	Oslo	Prague	Athens	Milan
Frying	2	2	2	
Frying with hood	3			
Candle burning	2	2		
Electrical cooker		3		
Oven		2		
Cooking (general)*		8	33	
Cooking (general) with hood			4	
Incense stick		2		
Aroma lamb		2		
Vacuum cleaner	1			
Smoking	1	2		2
Hair spray		2		1
Hair dryer		5		
Spray deodorant			2	
Air purifier			3	
Total time with indoor activities (min)	245	810	980	30
Percentage of time with indoor activities relative to total monitoring time	0.015%	0.040%	0.022%	0.002%

* Cooking (general) corresponds to the simultaneous use of more than one electrical devices in time periods less than 15 minutes.

day concentration variation. Furthermore, PN concentrations can exhibit strong spatial variations within the city limits according to the impact of traffic and to local pollution sources (Lianou *et al.*, 2007; Mejia *et al.*, 2008). Therefore, the outdoor PN concentrations will be mainly used to evaluate the impact of the outdoor environment to indoor air quality of the examined measurements' sites, since the infiltration of outdoor pollutants is strongly dependant on building characteristics.

The measured outdoor aerosol PN concentrations in the four cities presented significant differences. In order for the results to be comparable, the values in the size range 20–400 nm are presented, which was common in all campaigns (Table 2). These differences can be attributed to nearby pollution sources and to the elevation of the measurement sites from the ground (the office in Milan was in the ninth floor and the apartments in Athens were all in the fourth floor). The measurement sites in Prague and Oslo were at the suburbs of the city, while the sites in Milan and Athens were located at the city centre. The local meteorological conditions during the measurement periods also have a factor on the concentrations that reflect the dispersion and boundary layer depth.

Median PN concentrations in Milan and Athens for the 20–400 nm particles (13×10^3 # cm^{-3} and 23×10^3 # cm^{-3}) were more than four times and seven times higher respectively, compared to the corresponding value in Oslo (3×10^3 # cm^{-3}). The same applied for the average PN concentrations (Table 2). The site in Prague presented the lowest concentration levels (median: 2×10^3 # cm^{-3}) since it was located inside the Institute's campus and it was not influenced by major pollution sources (although the measurements were conducted during the cold season, emissions from domestic heating did not seem to affect outdoor PN concentrations significantly). Three times higher median value (7.3×10^3 # cm^{-3}) was reported by Borsos *et al.* (2012) for one year PN measurements in the same area of Prague, but there is almost 10 years difference between the two measurements and one year lasting campaign includes also periods heavily influenced by domestic heating and stronger traffic influence than in the case of a short campaign. On the other hand, the median outdoor PN concentrations in the three Athens' sites presented higher values mainly due to the influence of the emissions of the car fleet through the whole year and to the emissions from domestic heating during the cold season (Kavouras *et al.*, 2006). Increased PN levels in the Athens and Milan metropolitan areas were also reported in the literature. Lianou *et al.* (2011) measured median PN concentration values in Athens higher than Helsinki, Amsterdam and Birmingham. Lonati *et al.* (2011) conducted aerosol size distribution measurements for 22 days during the cold season at an urban background site in Milan and found median PN concentrations 1.87×10^4 for ultrafine particles and 4.89×10^3 for particles with mobility diameters between 100 nm and 1000 nm. Generally, following the work of Putaud *et al.* (2010), who presented PN concentration levels in 29 European sites, the particle load in Oslo and Prague resembles the situation in near city background sites like Ispra (Italy) and Melpitz (Germany), while PN values in Milan and Athens are closer to those of traffic influenced urban sites like Belfast, Glasgow and Barcelona.

Table 3 presents median indoor and outdoor number concentrations in four different size ranges. The 20–50 nm outdoor particles represented about 40% of total particles in Oslo, Milan and about 54% of total particles in Athens (Table 3(a)). Emissions in the 20–50 nm size range were 4.3 times higher in Milan and 9.8 times higher in Athens than in Oslo which clearly depicts the effect of the car fleet to Milan and Athens sites. It must be noted that the measurement height affects the evolution of the size distribution. Usually smaller particles emitted at the surface level are lost when mixed with clean air while transported vertically and probably the nano particles number concentrations would be even higher at the ground level in Milan and Athens. PN concentrations in the 200–400 nm size range were also higher in Athens and Milan, but their values were three times higher than Oslo. This fact corresponds to lower emissions of particles and particle precursors in this size range but also to reduced effects of the measurements' height to these particles. The absence of major pollution sources in the Prague measurement spot (emissions from diesel engines) resulted in lower outdoor number concentrations in the 20–50 nm size range.

Indoor Particle Number Concentrations

Indoor-to-Outdoor Relationship

Indoor number concentrations were lower than the outdoors (Table 2), with the exception of Prague case, where the indoor and outdoor median values were very close. In absence of indoor sources, the indoor number concentration

Table 2. Descriptive statistics of outdoor and indoor particle number concentrations (size range: 20–400 nm) in the four cities.

		Average ($\times 10^3$)	Std ($\times 10^3$)	Min ($\times 10^3$)	25% ($\times 10^3$)	Median ($\times 10^3$)	75% ($\times 10^3$)	Max ($\times 10^3$)
Oslo	Outdoor	3.79	2.11	0.68	2.26	3.22	4.69	16.04
	Indoor	3.37	10.19	0.69	1.34	1.78	2.52	174.99
Milan	Outdoor	13.95	6.36	2.97	8.94	13.00	18.13	48.37
	Indoor	9.30	10.90	3.04	5.68	7.71	10.12	211.37
Prague	Outdoor	2.90	3.59	0.22	1.03	1.75	3.33	54.69
	Indoor	5.99	20.44	0.23	0.90	1.64	3.10	328.71
Athens	Outdoor	29.47	24.93	2.90	13.89	22.93	35.49	231.29
	Indoor	24.21	31.17	0.97	10.72	16.06	25.61	274.18

Table 3. Median Outdoor (a) and Indoor (b) particle number concentrations in four different size fractions for each city.

(a) Outdoor number concentration ($\times 10^3$ # cm^{-3})

Size range (nm)	20–50	50–100	100–200	200–400
Oslo	1.32	0.91	0.63	0.29
Prague	0.44	0.49	0.53	0.27
Milan	5.70	3.82	2.41	0.86
Athens	12.02	6.26	2.92	0.91

(b) Indoor number concentration ($\times 10^3$ # cm^{-3})

Size range (nm)	20–50	50–100	100–200	200–400
Oslo	0.60	0.52	0.43	0.20
Prague	0.35	0.49	0.49	0.23
Milan	2.41	2.60	1.84	0.66
Athens	7.29	4.78	2.45	0.83

in naturally ventilated houses is determined by the outdoor aerosol concentration (Kearney *et al.*, 2011; Morawska *et al.*, 2001). In Prague, the low outdoor concentration levels magnified the impact of indoor sources to the indoor number concentration levels and resulted in high standard deviation values. In Oslo, Milan and Athens the median indoor PN concentrations were lower from the outdoors by 45%, 40% and 30% respectively. In Athens, the windows were open for longer time periods than in the other cities, which resulted in closer indoor and outdoor median PN concentrations.

The percentage of indoor particles with diameter 20–50 nm to the total number concentration was lower than the corresponding outdoor percentage in Oslo, Athens and Milan (Table 3(b)) and ranged between 31% in Milan to 45% in Athens. In Prague the percentage of indoor particles in the above size range was 21%, which was slightly lower than the outdoor percentage (25%). This can be attributed to the lack of major outdoor pollution sources and furthermore to indoor emissions throughout the day. The apartment in Prague was occupied all the time, which resulted in the formation of steady conditions between the indoor and outdoor environment. The median indoor number concentration was reaching the value of the corresponding median outdoor concentration as the particle's diameters were increasing in each size range and presented almost equally values for the 200–400 nm particles in Oslo, Prague and Athens. Generally, 20–50 nm particles penetrate less into the indoor environment than particles with diameters in the range 100–200 nm since diffusional losses during penetration are more efficient as the particle size gets smaller. This is more clearly shown in the Milan case where there were not any indoor sources, besides smoking of two cigarettes and use of hair spray (one time) during the measurement campaign. Similar results were presented by Hussein *et al.* (2005) who conducted indoor/outdoor measurements in a family house in Espoo (Finland) and reported increasing penetration efficiency with the increasing mobility diameters of particles up to 400 nm. Furthermore, studies in chambers showed that particles < 100 nm present higher deposition rates by Brownian diffusion in cracks and gaps of the building skeleton and therefore these particles cannot easily enter to the indoor environment (Jeng *et al.*, 2001; Mosley *et al.*, 2001). The effect of the specific building characteristics to the particle's penetration indoors can be seen in Milan, where the outdoor concentration was about 20% higher than the indoor concentration for the 200–400 nm particles. Since there were limited indoor emissions in Milan during this campaign, it is concluded that these particles can penetrate indoors more difficult than in the other sites where the indoor and outdoor PN concentrations for the 200–400 nm particles were almost the same.

Effect of Indoor Sources on the I/O Ratios

Indoor to outdoor ratios (I/O) were calculated for four selected size ranges (< 50 nm, 50–100 nm, 100–200 nm and > 200 nm in order to use all the available data in each site). Furthermore, the I/O ratios were sorted according to the influence of emissions due to indoor activities. Results are presented in Figs. 1(a)–1(c). In Prague, the central heating was always on during periods without indoor sources. The I/O ratios are size dependent (Morawska *et al.*, 2001; Hussein *et al.*, 2005) and in absence of any indoor activity they presented the lowest values for < 50 nm particles and the ratios increased with particle size.

The presence of people in the unoccupied apartment and in the office (Oslo and Milan respectively) resulted in increased I/O ratios compared to the corresponding I/O ratio values for periods without any indoor activities. The increase was observed even for particles < 50 nm, since some of the activities involved opening of windows. Generally, windows were open in Athens, Oslo and Milan for 1–2 hours each day with the difference that in Athens the time periods with open windows were not continuous and were divided in time intervals during the day. In Prague the windows were most of the time closed, probably due to lower outdoor temperatures (the apartment was occupied during the measurement period). The I/O ratios in Athens and Prague were not influenced by the presence of people and presented similar values also for periods without indoor activities. It must be noted that in these sites, the presence of people was not limited only to certain fixed hours each day but was varied according to the residents' schedule. Indoor sources (cooking, cigarette smoking, vacuuming, etc.) led to higher indoor concentrations compared to the

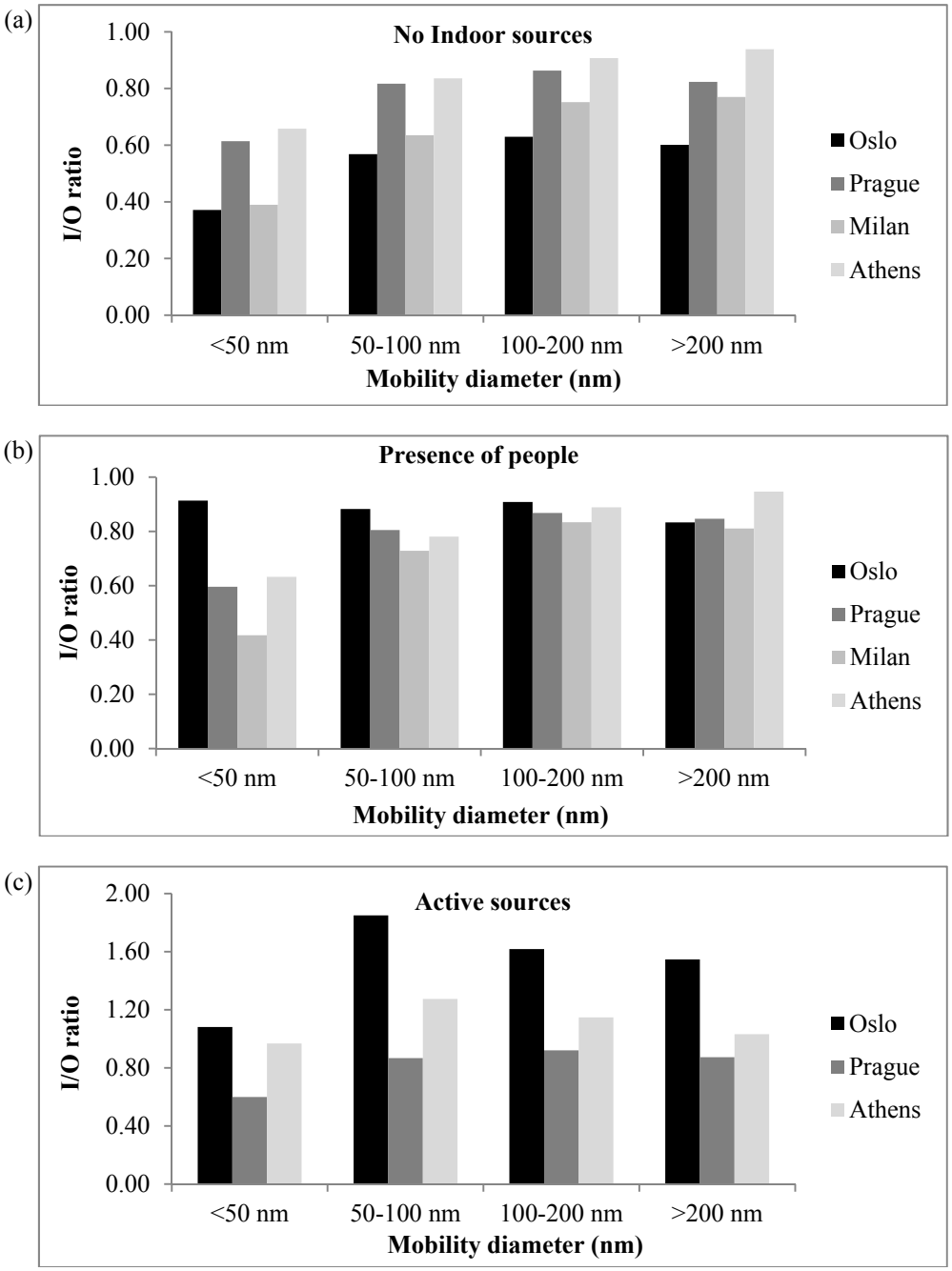

Fig. 1. I/O ratios for the four cities for time periods without any indoor activities (a), presence of people in the measurement sites (b) and active indoor sources (c). Milan is not presented in Figure 1c since only three indoor sources were active during the whole measurement campaign.

outdoors for all size ranges. The I/O ratios were lower than one only for the particles < 50 nm during periods with indoor sources, although some of the recorded activities (e.g., cooking and candle burning) emit mostly nano particles (Afshari *et al.*, 2005; Buonanno *et al.*, 2009; Glytsos *et al.*, 2010). In these cases, the emitted particles were collected in the instruments in larger sizes due to the coagulation and condensation processes. Furthermore, smaller particles present higher deposition rates on indoor surfaces while particles in the accumulation mode (size range 200 nm to 400 nm) are too big to diffuse on surfaces through Brownian motion and too small to be removed via gravitational settling with higher rate (Hinds, 1999). Even in cases when the increase of the concentration was not so intense and was limited only to bigger particles (e.g., hair spray and vacuum cleaning), the accumulation mode dynamics and the higher deposition rates of smaller particles also influenced indoor particles of outdoor origin and the I/O ratios with active sources were not significantly affected.

In order to evaluate the contribution of indoor emissions and outdoor particles penetration to the indoor concentration, the diurnal variation of the number concentration was

computed for each site both for indoor and outdoor aerosols. In Oslo and Milan, only simulated indoor activities were performed, mostly until noon and during the afternoon and the evening the sites were unoccupied. In Athens and Prague, the people living in the residences recorded in detail the indoor activities during their presence in the indoor environments. The Pearson correlation coefficient between indoor and outdoor concentration was computed and selected results for Oslo and Prague are presented in Figs. 2(a) and 2(b). It is clearly shown that indoor and outdoor particles are correlated in absence of indoor sources, but the magnitude of correlation is dependant on the specific building characteristics and on the meteorological conditions that affect the outdoor particles infiltration into the indoor environment. Furthermore, the low correlation of indoor and outdoor particle number concentrations between 15:00 and 21:00 could be attributed to the fact that the measurement staff was leaving from the apartment between 14:00 and 15:00 and windows and doors remained closed for the rest of the day (the air exchange between the measurement's room and the other rooms of the house was also limited). This resulted probably in lower deposition rates of the derived particles from the simulated activities conducted during the morning and to slower renewal of the indoor air. Pearson coefficients during non activities periods presented values very close to 1 for particles bigger than 100 nm. The correlation weakened as the particle size reduced due to reduced penetration efficiency into the indoor environment. On the other hand, indoor and outdoor particles were not strongly correlated during periods with presence of people and no correlation was observed (Pearson correlation coefficients presented values close to zero) during periods with active indoor sources. In these periods the indoor concentration was defined mainly by indoor emissions.

Modal Structure of Indoor and Outdoor Aerosols

The outdoor median size distributions (Fig. 3) showed similar characteristics in Oslo and Milan regardless the absolute values of the median PN concentrations and the position of the Aitken mode (~20 nm for Oslo and ~40 nm for Milan). Both size distributions were bimodal with lower GMDs in Oslo (16 nm and 73 nm) compared to Milan (37 nm and 129 nm). The first mode indicated the presence of Aitken particles, probably as a result of emissions from traffic and domestic heating. Emissions of nano particles were more intense in Milan, as the first mode was the dominant mode in the size distribution. The effect of smaller particles in the shape of the median size distribution was even greater in Athens, which was bimodal, with GMDs at 29 nm and below 10 nm. On the other hand, the Prague median distribution was unimodal with GMD at 79 nm, showing minor effect from vehicular fleet emissions. The indoor median size distributions were similar in the shape with the outdoors for Oslo and Milan, suggesting that in these sites the penetration of outdoor aerosol determined the characteristics of indoor

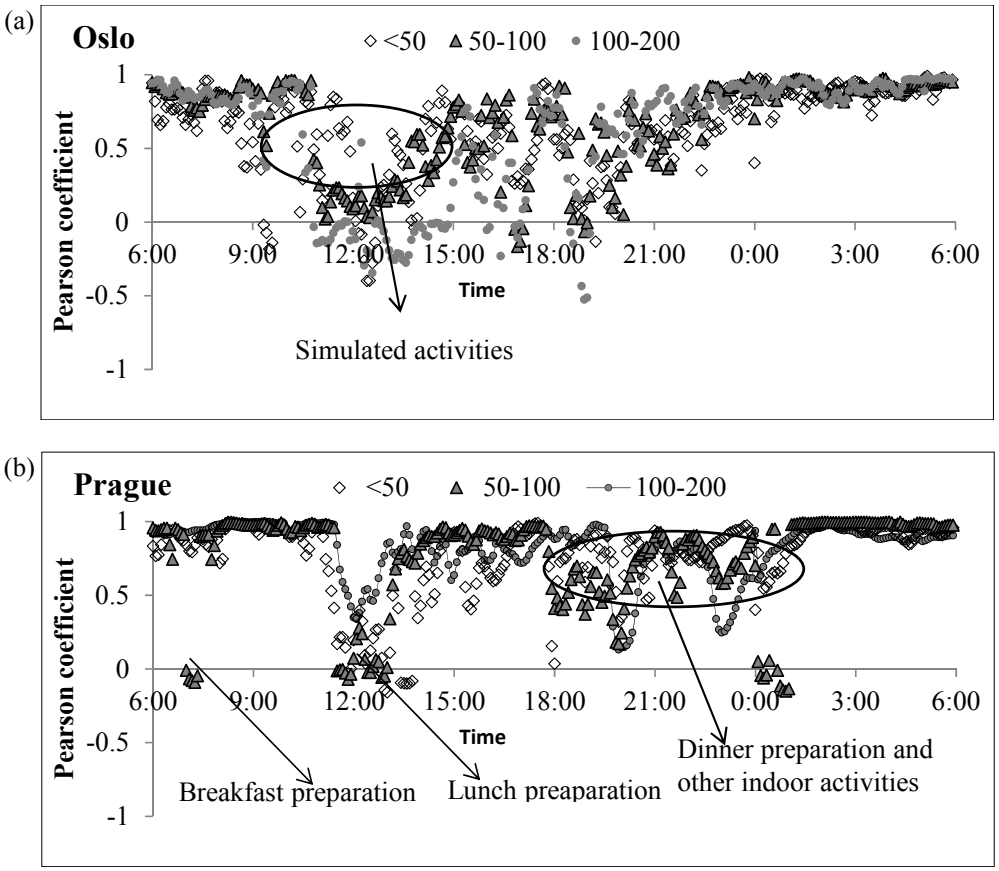

Fig. 2. Diurnal variation of Pearson coefficient for Indoor/Outdoor PN concentrations for Oslo (a) and Prague (b).

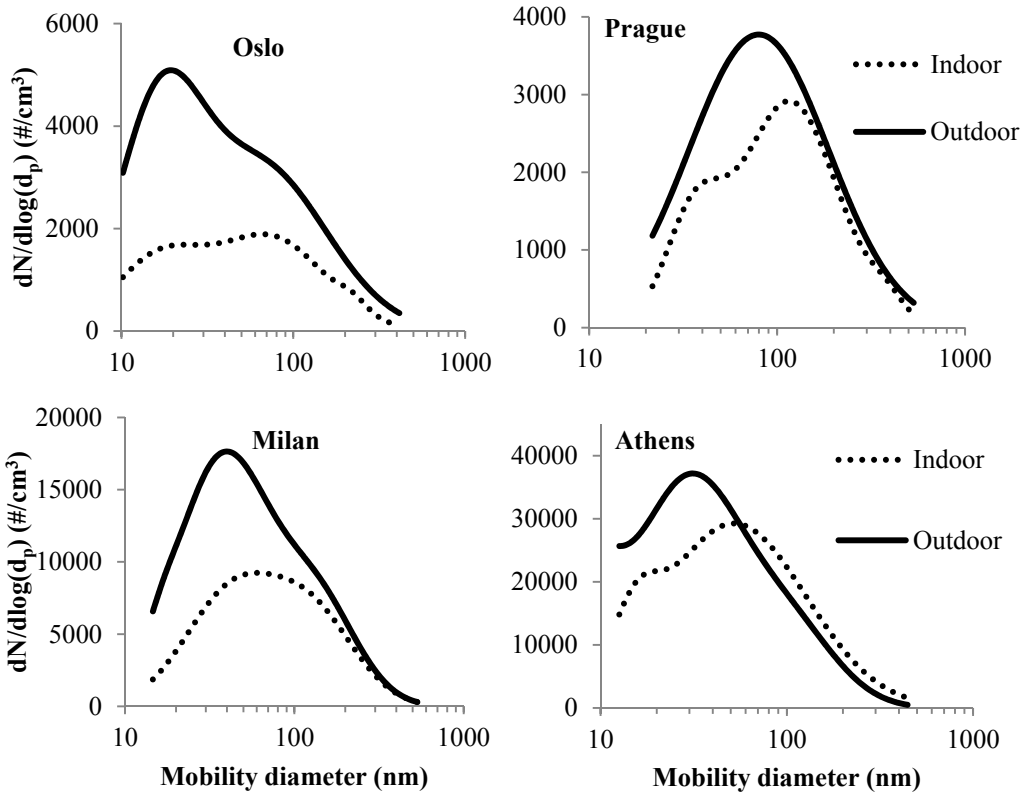

Fig. 3. Median size distributions for Outdoor/Indoor aerosol in the four cities.

particles. It must be noted that the house in Oslo and the office in Milan were unoccupied during the night in contrast to Athens and Prague. Furthermore, in Milan there were limited indoor activities besides possible emissions due to the presence of people. The median indoor size distribution in Prague was bimodal and the first mode (GMD at 36 nm) can be attributed mainly to indoor activities. The indoor air in Athens was affected both from incoming outdoor particles and from indoor emissions as it can be seen from the median indoor size distribution.

The derived size distributions from the PN concentration measurements were analysed and their characteristics are summarized in Tables 4 and 5. The time periods with presence of people in the indoor environment, without emissions from any active sources (cooking, cigarette smoking, etc.), were included in periods without indoor activities (Table 4(b)). Moreover, periods with indoor activities were considered equivalent to time intervals between the start of the emissions until one hour after the end of it, as long as the indoor concentration was higher than the background concentration.

The results showed that without indoor activities the percentages of unimodal size distributions were almost the same for indoors and outdoors (Table 5) for each measurement site with the exception of Athens, where the percentage of the outdoor unimodal size distributions was higher (35% for the indoor unimodal size distributions and 45% for the corresponding outdoor size distributions). PN concentrations were generally lower indoors in periods without indoor activities (Tables 4(a) and 4(c)). Significant differences were depicted for the 2nd and the 3rd modes for the bimodal and trimodal size distributions in all sites (Tables 4(a) and 4(c)) and the diversity in the size distribution characteristics was more evident in the Prague and Athens data, indicating probable emissions due to activities associated to the presence of people.

Emissions from indoor sources led in elevated PN concentrations in the 1st mode (Table 4(b)), as a result of particle emissions with diameters below 50 nm. Also the GMDs in Oslo and Prague for unimodal size distributions were different under the influence of indoor sources, showing that indoor sources emit particles in specific size ranges according to the kind of the activity. Unimodal size distributions during emissions from controlled indoor activities in a laboratory room were reported by Glytsos *et al.* (2010).

Furthermore, the median GMDs and PN concentrations were computed for Aitken and Accumulation modes particles (Table 6). A fraction of the particles in the Nucleation modes could not be detected by the instrumentation used in measurement campaigns and therefore these modes were excluded from the analysis of the collected data. Without indoor activities, GMDs for the indoor and outdoor particles were almost the same, with higher PN concentrations outdoors. During periods with indoor activities, the GMDs of the Aitken mode in Oslo, Prague were shifted towards higher diameters and PN concentrations were higher indoors both for Aitken and Accumulation modes. GMD values of the indoor size distributions in the Accumulation mode in Athens and Oslo were less affected by indoor sources and

Table 4. Median values of GMDs, number concentration and geometric standard deviation for outdoor aerosol (a) and for indoor aerosol sorted according to time period without indoor activities (b) and with indoor activities (c). The numbers 1, 2, 3 refer to the corresponding mode (1 for 1st mode, etc.)

(a) Median values for outdoor aerosol

		GMD [nm]			Number concentration [# cm^{-3}]			GSD		
		1	2	3	1	2	3	1	2	3
Unimodal	Prague	107			1800			2.1		
	Oslo	45			5700			2.2		
	Milan	47			17000			2.2		
	Athens	32.7			52000			2.3		
Bimodal	Prague	31	119		470	1500		1.5	2.0	
	Oslo	20	97		2500	2000		1.7	1.9	
	Milan	33	121		9400	5900		1.8	1.8	
	Athens	24	108		30000	12100		2.2	1.8	
Trimodal	Prague	29	91	264.5	450	800	260	1.5	1.6	1.6
	Oslo	18	70	212.9	1700	1300	720	1.7	1.6	1.5
	Milan	18	45	141.1	3600	5100	3700	1.7	1.6	1.7
	Athens	12	36	110.9	19600	14200	8210	2.0	1.7	1.8

(b) Median values for time periods without indoor activities

		GMD [nm]			Number concentration [# cm^{-3}]			Geometric Standard deviation		
		1	2	3	1	2	3	1	2	3
Unimodal	Prague	120			1200			2.0		
	Oslo	57			2200			2.2		
	Milan	74			7300			2.0		
	Athens	36			22300			2.9		
Bimodal	Prague	34	132		460	800		1.7	1.9	
	Oslo	23	91		1070	1400		1.5	1.5	
	Milan	41	128		3200	4600		1.7	1.8	
	Athens	29	130		14400	3500		2.2	1.6	
Trimodal	Prague	26	111	339	220	740	150	1.5	1.7	1.4
	Oslo	14	49	156	750	830	580	1.6	1.5	1.6
	Milan	22	54	183	140	3700	1700	1.6	1.7	1.6
	Athens	10	54	256	15500	17300	4000	2.1	1.8	2.0

(c) Median values for periods with indoor activities

		GMD [nm]			Number concentration [# cm^{-3}]			Geometric Standard deviation		
		1	2	3	1	2	3	1	2	3
Unimodal	Prague	85			28000			1.7		
	Oslo	85			42200			1.7		
	Athens	35			37000			3.0		
Bimodal	Prague	45	182		9700	1900		1.5	1.6	
	Oslo	20	101		5000	2900		1.5	1.6	
	Athens	27	123		20500	4500		2.2	1.6	
Trimodal	Prague	31	105	369	5900	2000	450	1.50	1.6	1.6
	Oslo	6	21	97	1850	1900	4950	1.7	1.4	1.7
	Athens	11	45	269	33500	48000	13000	2.0	1.8	2.1

their characteristics were related to the characteristics of the outdoor size distributions. In Prague the case was different, and the GMD of the accumulation mode was increased. In Athens, the emissions from indoor sources also resulted in elevated indoor concentrations, compared to periods without indoor activities, but the GMDs both in Aitken and accumulation modes presented similar values in all indoor scenarios. In addition, the values of the GMDs in the indoor environment during periods with active indoor sources were very close to the corresponding GMDs in the outdoor environment, probably as a result of higher air penetration from outdoors. The above results indicate that indoor emissions can alter in many cases significantly the characteristics of indoor air and their role must be thoroughly evaluated in indoor air quality studies.

Characteristics of Specific Indoor Activities

Several indoor activities were registered during the

Table 5. Percentage of Unimodal, Bimodal and Trimodal size distributions in each city.

		Percentages (%)		
		Unimodal	Bimodal	Trimodal
Oslo	Outdoor	5.70	63.50	30.80
	Indoor (No active sources)	5.10	43.90	51.00
	Indoor (Active sources)	30.85	43.00	26.15
Prague	Outdoor	22.80	59.90	17.30
	Indoor (No active sources)	26.70	49.80	23.50
	Indoor (Active sources)	28.20	53.20	18.60
Milan	Outdoor	23.60	60.00	16.40
	Indoor (No active sources)	21.45	56.95	21.60
	Indoor (Active sources)			
Athens	Outdoor	44.20	22.90	30.90
	Indoor (No active sources)	35.00	56.50	8.50
	Indoor (Active sources)	36.00	53.00	11.00

Table 6. Modal structure of Aitken and Accumulation modes of the collected data in the four cities.

		Aitken Mode			Accumulation Mode		
		GMD (nm)	PN Conc. (# cm^{-3})	GSD	GMD (nm)	PN Conc. (# cm^{-3})	GSD
Oslo	Outdoor	27.8	2208	1.64	163.4	1004	1.7
	Indoor (No active sources)	31.7	1045	1.66	153.7	637	1.6
	Indoor (Active sources)	48.1	12031	1.63	150.7	5532	1.6
Prague	Outdoor	33.7	559	1.51	129.0	467	1.6
	Indoor (No active sources)	37.0	359	1.61	131.2	847	1.8
	Indoor (Active sources)	53.9	5363	1.64	182.2	1506	1.5
Milan	Outdoor	37.6	8820	1.84	136.2	4718	1.7
	Indoor (No active sources)	44.6	4059	1.72	137.1	4216	1.8
Athens	Outdoor	36	19880	2.05	134	2963	1.61
	Indoor (No active sources)	39	17143	2.19	145	2756	1.56
	Indoor (Active sources)	37	23226	2.20	132	3670	1.62

measurement campaigns in the four cities. A detailed analysis of the specific characteristics (emission rates, size distributions, deposition rates, etc.) of each indoor source is beyond the objectives of this work. Furthermore, many of the activities were happening simultaneously or they were overlapping for some period of time and thus it was very difficult to distinguish the effects of each source to the indoor PN size distribution spectra. Nevertheless, two activities were selected for presentation in order to highlight possible differences and similarities between emissions in different indoor environments and under different indoor conditions.

Frying (Oslo, Athens, Prague)

On all occasions frying was performed on an electrical stove and no other activities took place one hour after the end of the emission periods. Only one person was present in the kitchen in Oslo, two persons were in the kitchen in Prague and three persons were at the apartment in Athens. The hoods were not working during frying in all the occasions. Contour plots of frying in each measurement site are presented as supplementary material. The PN concentrations reached their maximum values at the end of the emissions and they were of the same order of magnitude (1.71×10^5 # cm^{-3}, 2.6×10^5 # cm^{-3} and 1.35×10^5 # cm^{-3} in Oslo, Athens and Prague respectively). UFP constituted 70% of submicron particles in the Oslo case, 65% of submicron particles in Athens, while the corresponding percentage of UFP in Prague was 90%. Size distributions were unimodal during and after the end of the emissions as it is depicted in Fig. 4 for Prague and Oslo. The corresponding GMDs ranged between 70 nm and 105 nm in Oslo (90 nm at the end of emissions), between 30 nm and 55 nm in Prague (38 nm at the end of emissions) and between 80 and 100 nm in Athens (close to 90 nm at the end of emissions). These differences cannot be attributed to specific factors since the number concentration and the size distribution of the emitted particles during frying are strongly dependent on various parameters such as the type of frying (Dennekamp *et al.*, 2001; See and Balasubramanian, 2006), the type of the food that was used (Li *et al.*, 1993) and the temperature of frying (Yeung and To, 2008). The emitted particles experienced strong coagulation effects which resulted in the shift of the GMDs towards higher values. This phenomenon continued after the termination of sources (Figs. 4 and 5).

Smoking (Prague, Oslo and Milan)

Tobacco smoking produced mainly particles in the size range 50 nm–200 nm (Figs. S4–S8 in supplementary material). In all the cases, the percentage of particles in the

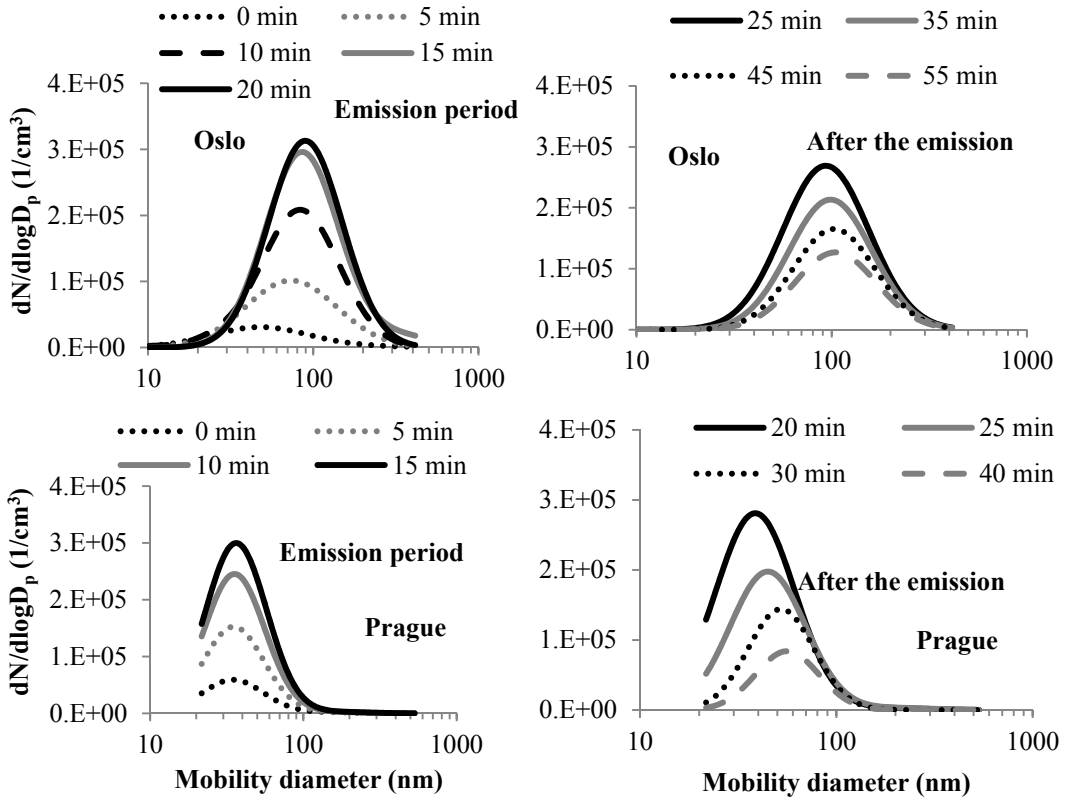

Fig. 4. Median size distributions during and after the emissions from frying in Oslo (upper figures) and Prague (lower figures).

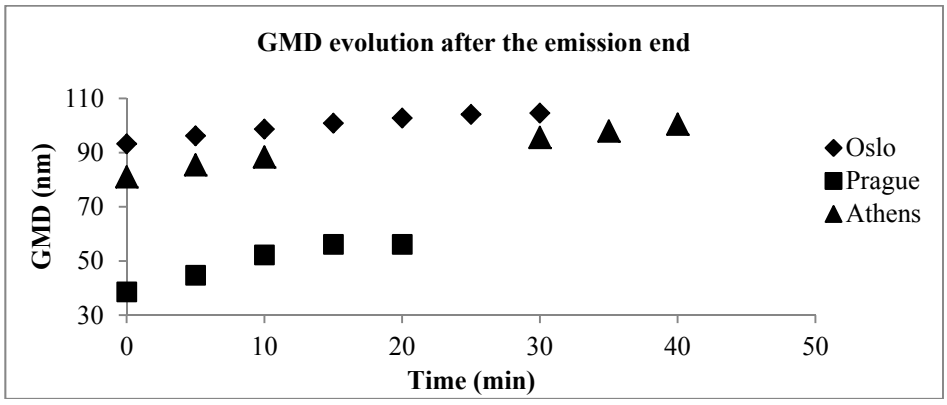

Fig. 5. GMD evolution after the end of emissions from frying in Oslo, Athens and Prague.

above size range was more than 70% of total submicrometer particles, from the beginning of the emissions until one hour after the cigarettes were extinguished. Maximum PN concentrations ranged between 3.0×10^4 # cm^{-3} (Prague) and 1.7×10^5 # cm^{-3} (Milan). The GMDs at the time of maximum concentrations varied between 81 nm to 113 nm (Fig. 6) due to differences in the type of cigarette, in the smoking patterns and in the characteristics of the indoor environments.

CONCLUSIONS

Indoor/outdoor aerosol size distribution measurements have been performed in four European cities (Oslo – Norway, Prague – Czech Republic, Milan – Italy and Athens – Greece) to analyse their characteristics and to determine the differences between the indoor and outdoor size distributions. The measurement sites were naturally ventilated and indoor activities were performed by the permanent residents or by the technical staff responsible for the instrumentation. All activities were registered in time-activity diaries. Regarding the outdoor aerosol, the PN concentrations and modal structure characteristics presented significant differences among the measurements sites, mainly due to their locations, since the sites in Athens and Milan were in the city centers and the sites in Oslo and

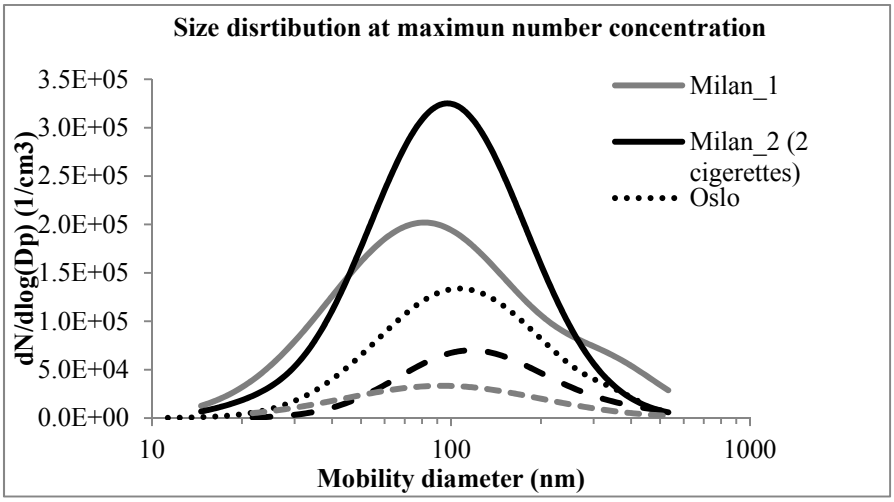

Fig. 6. Size distributions at the time of maximum number concentrations (end of emissions) during smoking in Milan (two experiments), Oslo and Prague (two experiments).

Prague were in the suburbs. In addition, the measurements height affected the outdoor PN concentrations for smaller particles and reduced the available for penetration outdoor particles in the size range 20–50 nm. Particles with diameters lower than 50 nm represented more than 50% of total particles in Oslo, Milan and Athens, while the absence of major pollution sources in Prague resulted in lower outdoor number concentrations in the < 50 nm size range. In absence of indoor sources, PN concentrations indoors were lower than outdoors and the indoor concentration followed the variations of the outdoor. The percentage of < 50 nm indoors was lower than outdoors in the Oslo, Athens and Milan sites indicating, as expected, that nano particles penetrate more difficult in the indoor environment than particles in the 100–200 nm size range. The 200–400 nm particles presented similar indoor and outdoor concentrations, with the exception of Milan, where the indoor PN concentration in this size range was lower than the outdoor. This fact can be associated to the specific characteristics of the building envelope in Milan.

The I/O ratios presented the lowest values for < 50 nm particles ratios and their values increased with particle size. The presence of people led to higher indoor concentrations and increased I/O ratios while during emissions from indoor sources I/O ratios presented values higher than one. The presence of people in Oslo was directly associated with opening the windows for 1–2 hours and therefore the I/O ratios were close to one. The I/O ratios in Athens did not present significant differences for the > 100 nm particles, since opening the windows was not limited to time periods with presence of people. In Prague, the windows were closed all the time, which resulted in an establishment of steady state conditions between the indoor and outdoor environment and similar I/O ratios with and without the presence of people in the apartment. Indoor and outdoor concentrations were strongly correlated in time periods without indoor activities and the correlation decreased as the particle size reduced. On the other hand, indoor and outdoor particles were not strongly correlated during periods with presence of people and no correlation was observed during periods with active indoor sources, which demonstrates the already known effects of indoor sources to the indoor particle number concentrations.

Indoor activities affected the indoor modal structure and resulted in the strong presence of Aitken mode particles in the indoor environment. The percentage of unimodal size distributions increased during indoor emissions indicating the presence of particles in specific size ranges according to the indoor activity and usually overlapping the multi-modal structure of the background aerosol. Therefore, this study confirms that indoor size distributions cannot derive from the outdoor only and the emission characteristics of indoor sources must be considered during indoor air quality studies.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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