Global NEST Journal, Vol 12, No 1, pp 12-19, 2010

INDOOR / OUTDOOR PM LEVELS AND EC SURROGATE, AT TYPICAL MICROENVIRONMENTS IN THE ATHENS AREA

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Received: 21/12/09
Accepted: 09/02/10

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ABSTRACT

The aim of the present work was to characterize particulate matter (PM) and elemental carbon (EC) indoor and outdoor concentration levels in the Athens area and to examine the contribution of ambient air to the observed indoor levels.

24-hr simultaneous indoor and outdoor PM_{10} and $PM_{2.5}$ measurements were conducted at a central (CR) and a suburban (SR) residence, and at an office in the commercial centre of Athens (CO), during cold and warm period of 2006. The absorption coefficient (α) was measured on the collected filters, as a surrogate for EC concentration levels.

Ambient PM levels were very high at both central sites and significant at SR (mean 24-hr PM_{10}: 87.4, 50.3 and 87.3 μg m^{-3} and $PM_{2.5}$: 50.7, 20.2 and 42.8 μg m^{-3} at CR, SR and CO). The measured absorption coefficient values were very high at CR and CO for both size fractions.

Indoor PM concentration and absorption coefficient values were lower than the respective outdoor ones, but still significant at the two central sites.

Very good correlations were observed between indoor and outdoor data (especially for absorption coefficient values), indicating a large contribution of the ambient atmosphere to the indoor levels, more pronounced in finer particles.

KEYWORDS: Indoor / outdoor levels, $PM_{10}/PM_{2.5}$, Absorption coefficient, Elemental carbon, Indoor particles of outdoor origin.

INTRODUCTION

Numerous epidemiological studies have documented that current day levels of particulate matter (PM) air pollution are associated with adverse health effects, including increased risks of morbidity and mortality, mainly due to respiratory and cardio-vascular diseases (Pope and Dockery, 2006; Pope *et al*., 2002). Research interest is directed towards densely populated urban centres, where significant ambient PM concentrations have been observed. The main source of particulate air pollution in these areas is vehicular traffic. An important component of traffic-related PM is elemental carbon (EC). EC is the dominant light-absorbing substance in the atmosphere. It is composed primarily of carbon-based chemical by-products formed from incompletely combusted or un-combusted hydrocarbon-based fuels.

Event though epidemiological studies examine association of health effects with ambient concentration levels, the scientific interest has lately turned towards the indoor microenvironment. Urban populations spend about 90 % of the time indoors, with the

[†] Prof. Spyrellis suddenly passed away. We would like to dedicate this work to his memory.

residence and work microenvironments being the prevailing ones according to the duration of occupancy (Adgate *et al*., 2002; Brauer *et al*., 2000). It has become apparent, thus, that indoor concentration levels may contribute significantly to total personal exposure, often exceeding the respective contribution of the ambient atmosphere. Therefore, associations between outdoor levels of air pollution and health outcomes may be plausible only if health relevant constituents of ambient air pollution efficiently penetrate indoors.

The aim of the present work was the characterization of PM (PM_{10} and $PM_{2.5}$) and elemental carbon (EC) indoor and outdoor concentration levels in the Athens area, at two typical microenvironments: the residential and office microenvironment. Moreover, indoor and outdoor data were analyzed in order to examine the contribution of ambient air to the observed indoor levels.

METHODS

Two residences, one in a central area of Athens (CR) and one in a quite suburb (SR), as well as an office in the commercial centre of Athens (CO), were selected for study. Both CR and CO are located in high-trafficked and densely populated areas, while SR is located in a neighborhood characterized by low traffic, no commercial activity and many green areas. All measurements took place during cold and warm period of 2006. Each site was studied for at least two weeks and up to one month. The under-study rooms were a bedroom at CR and SR, usually empty during the day, and a non-smoking office at CO. The specific rooms were selected as to present minimum indoor particle generation, in order to reveal the impact of ambient air to indoor concentration levels.

Simultaneous 24-hr indoor and outdoor measurements of PM_{10} and $PM_{2.5}$ mass concentration were conducted by the use of Harvard samplers, at a flow rate of 4 LPM. Both indoor and outdoor samplers were placed at a height of 1.0 – 1.5 m (breathing height). Indoor samplers were positioned in the centre of the under-study room, at some distance from walls and openings. Outdoor sampling took place at the terraces, outside the rooms.

The absorption coefficient (α) was measured on the collected PM_{10} and $PM_{2.5}$ filters by the use of a smoke stain reflectometer (Model EEL 043 D, Diffusion Systems, Ltd). Numerous studies have indicated that there is an excellent correlation between coefficient α and concentration of particulate EC, suggesting thus that absorbance of PM filters may be used as a surrogate for elemental carbon concentration levels (Adams *et al*., 2002; Kinney *et al*., 2000; Edwards *et al*., 1983).

RESULTS AND DISCUSSION

Meteorological conditions

Measurements were conducted during both cold and warm period. Cold period corresponded to the months November - April and warm period to the months May – October. Basic statistics for the daily values of main meteorological parameters are presented in Table 1.

Table 1. Basic statistics for the daily values of temperature (T), relative humidity (RH), wind speed (Ws) [mean ± standard deviation] and rainfall (Rf) [total rainfall height (number of days with rainfall episode)], during the measurement period

	T (ºC)	RH (%)	Ws (m s^{-1})	Rf (mm)
Cold period	12.0 ± 3.2	67.4 ± 9.6	1.5 ± 0.9	97.6 (24)
Warm period	20.0 ± 3.2	61.3 ± 14.2	2.3 ± 1.4	22.6 (9)

Outdoor measurements

The measured PM levels were comparable for cold and warm period. Mean daily outdoor PM concentration levels are presented in Table 2.

Ambient levels were very high at both central sites and significant at the suburban residential area, with the mean values in all three sites surpassing the E.U. 24-hr air quality standard for PM_{10} (50 μg m^{-3}). The fraction of days that exceedance of the 24-hr limit value was observed, was calculated equal to 84 % for CR, 57 % for SR and 100 % for CO. Regarding $PM_{2.5}$

concentration levels, at the two central sites mean concentrations were greater than 25 μg m^{-3}, which is the E.U. annual limit value for this size fraction, to be achieved during 2010 - 2020. If the measured concentrations are compared with the 24-hr limit value of 35 μg m^{-3}, proposed by the C.A.F.E. working group (2004), exceedances were observed during 69 % and 43 % of the days at CR and CO, respectively.

Table 2. Mean daily outdoor PM_{10} and $PM_{2.5}$ concentration levels [μg m^{-3}]

	PM_{10}		**$PM_{2.5}$**	
	Mean ± St. Dev.	**Range**	**Mean ± St. Dev.**	**Range**
CR	87.4 ± 28.5	37.2 – 134.7	50.7 ± 18.8	29.8 – 80.3
SR	50.3 ± 9.1	36.0 – 61.4	20.2 ± 5.7	13.9 – 26.3
CO	87.3 ± 28.1	55.9 – 161.1	42.8 ± 16.7	25.3 – 87.4

The value of the $PM_{2.5}$-to-PM_{10} concentration ratio reflected the main source affecting each site. Mean $PM_{2.5}/PM_{10}$ ratio was calculated equal to 0.72 and 0.70 at CR and CO, indicating a strong contribution of vehicular traffic, which produces mainly fine particles. The corresponding ratio at SR was much lower (equal to 0.42), indicative of stronger coarse particle sources, which may be attributed to the free land and green areas surrounding the under-study site.

The measured absorption coefficient (α) was high at the two central sites (CR and CO). Coefficient α may be used as surrogate for particulate elemental carbon (EC) concentration. It is therefore expected to present significant values in areas with high vehicular traffic, which is considered the main EC source in urban areas (Janssen *et al*., 2001). Basic statistics for the absorbance measurements on the filters collected outdoors are presented in Table 3.

Table 3. Mean daily outdoor absorption coefficient (α) values measured in PM_{10} and $PM_{2.5}$ filters [10^{-5} m^{-1}]

	α (PM_{10})		**α ($PM_{2.5}$)**	
	Mean ± St. Dev.	**Range**	**Mean ± St. Dev.**	**Range**
CR	5.9 ± 2.8	1.5 – 11.6	4.2 ± 1.7	2.1 – 7.7
SR	1.2 ± 0.2	0.8 – 1.6	1.0 ± 0.5	0.5 – 1.8
CO	4.2 ± 2.2	2.2 – 12.3	3.7 ± 1.5	2.3 – 7.0

The measured absorption coefficient values for $PM_{2.5}$ were similar to the ones reported in the literature. The respective mean daily values given in other studies, conducted in the city of Athens as well, are in the range $2.6 \cdot 10^{-5}$ – $3.5 \cdot 10^{-5}$ m^{-1} (Puustinen *et al*., 2007; Lai *et al*., 2006; Gotschi *et al*., 2002). Similar levels to the ones measured at the two central sites (CR and CO) have been also reported for Erfurt, Germany ($4.0 \cdot 10^{-5}$ m^{-1}) by Ruuskanen *et al*. (2001) and for Verona, Italy ($4.2 \cdot 10^{-5}$ m^{-1}) by Gotschi *et al*. (2005). These two studies present the highest values reported in the literature. The absorption coefficient values measured at the suburban site were compatible with the ones measured in other European cities, such as Helsinki, Finland, Oxford, U.K., Basel, Switzerland (Puustinen *et al*., 2007; Lai *et al*., 2006; Gotschi *et al*., 2002).

Relative results for PM_{10} are scarce. Fischer *et al*. (2000) reported PM_{10} absorption coefficient values for Amsterdam, Netherlands equal to $1.6 \cdot 10^{-5}$ m^{-1} (in a low-traffic site) and $3.0 \cdot 10^{-5}$ m^{-1} (in a heavy-traffic site). In another study in Huddersfield, U.K., the respective mean value was equal to $1.7 \cdot 10^{-5}$ m^{-1} (Kingham *et al*., 2000).

The absorption coefficient presented a very good correlation with mass concentration for both size fractions (r equal to 0.84 for PM_{10} and 0.80 for $PM_{2.5}$). Elemental carbon is expected to correspond mainly to fine particles (with an aerodynamic diameter of less than 1 μm) (Seinfeld & Pandis, 2006; Chow, 1995). Nevertheless, the high correlation coefficient between

PM_{10} and the respective absorption coefficient values indicated that a significant portion may be found in coarse particles as well.

Indoor measurements

The measured mean daily indoor PM_{10} and $PM_{2.5}$ concentration levels are presented in Table 4. Indoor PM_{10} and $PM_{2.5}$ concentrations were lower than the respective outdoor ones at all three sites. Nevertheless, significant levels were observed in the two central sites for both PM fractions. The fraction of days that exceedance of the 24-hr limit value for ambient PM_{10} concentration was observed, was calculated equal to 58 % for CR and 23 % for CO. In SR there was no exceedance. Regarding $PM_{2.5}$, concentration levels were greater than the 24-hr limit value of 35 μg m^{-3}, proposed by the C.A.F.E. working group (2004), during 31 % and 22 % of the days at CR and CO, respectively. Again, no exceedance was observed in SR.

Table 4. Mean daily indoor PM_{10} and $PM_{2.5}$ concentration levels [μg m^{-3}]

	PM_{10}		**$PM_{2.5}$**	
	Mean ± St. Dev.	**Range**	**Mean ± St. Dev.**	**Range**
CR	51.6 ± 15.2	24.9 – 79.4	31.9 ± 8.5	22.0 – 47.7
SR	20.5 ± 4.2	15.8 – 28.1	16.6 ± 3.6	12.8 – 21.0
CO	56.5 ± 28.1	28.6 – 185.5	37.4 ± 14.9	24.3 – 83.2

The absorption coefficient values measured on the indoor filters were slightly lower than the respective outdoor values (Table 5). Nevertheless, again there was a significant difference between the values measured at central sites in relation to the suburban site, confirming that traffic-related air pollution may greatly affect the indoor microenvironments (Wichmann *et al*., 2005). The absorption coefficient values presented a good correlation with PM mass for both size fractions (r equal to 0.73 for PM_{10} and 0.83 for $PM_{2.5}$).

Table 5. Mean daily indoor absorption coefficient (α) values measured in PM_{10} and $PM_{2.5}$ filters [10^{-5} m^{-1}]

	α (PM_{10})		**α ($PM_{2.5}$)**	
	Mean ± St. Dev.	**Range**	**Mean ± St. Dev.**	**Range**
CR	4.9 ± 2.5	1.0 – 9.7	3.1 ± 1.7	1.2 – 6.0
SR	0.9 ± 0.2	0.7 – 1.2	0.8 ± 0.4	0.4 – 1.4
CO	3.8 ± 1.8	2.2 – 11.3	3.3 ± 1.3	2.2 – 6.8

Indoor – outdoor relationships

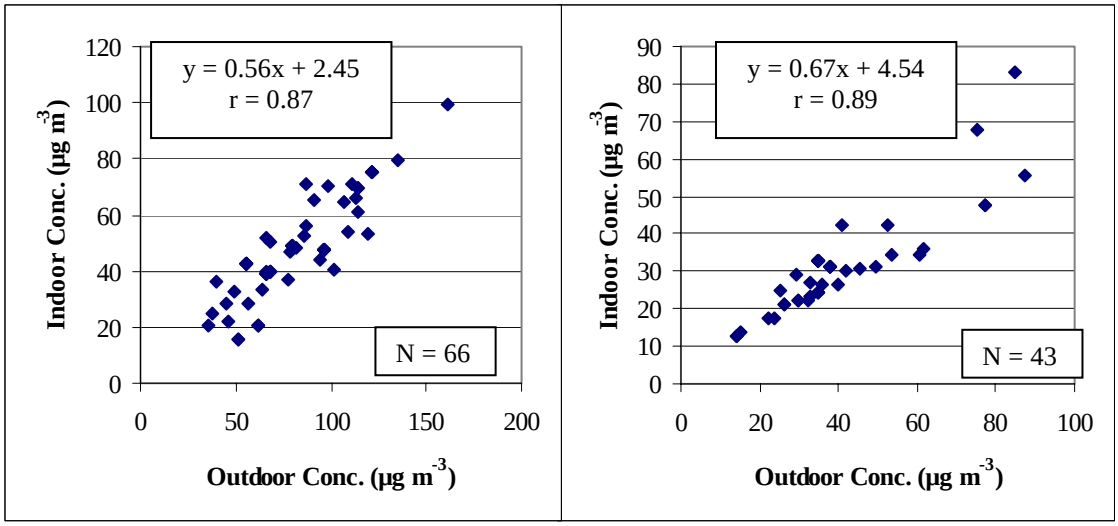
Indoor-to-outdoor concentration (I/O) ratios were below 1.00 at all sites and during all measurement days, indicating that there was no significant indoor source (Table 6). Thus, indoor concentration levels were expected to be mainly affected by the ambient atmosphere.

Table 6. Mean daily I/O ratios for PM_{10} and $PM_{2.5}$

	PM_{10}		**$PM_{2.5}$**	
	Mean ± St. Dev.	**Range**	**Mean ± St. Dev.**	**Range**
CR	0.61 ± 0.12	0.40 – 0.90	0.66 ± 0.09	0.50 – 0.83
SR	0.42 ± 0.14	0.31 – 0.63	0.84 ± 0.07	0.75 – 0.92
CO	0.61 ± 0.09	0.47 – 0.82	0.87 ± 0.11	0.64 – 0.99

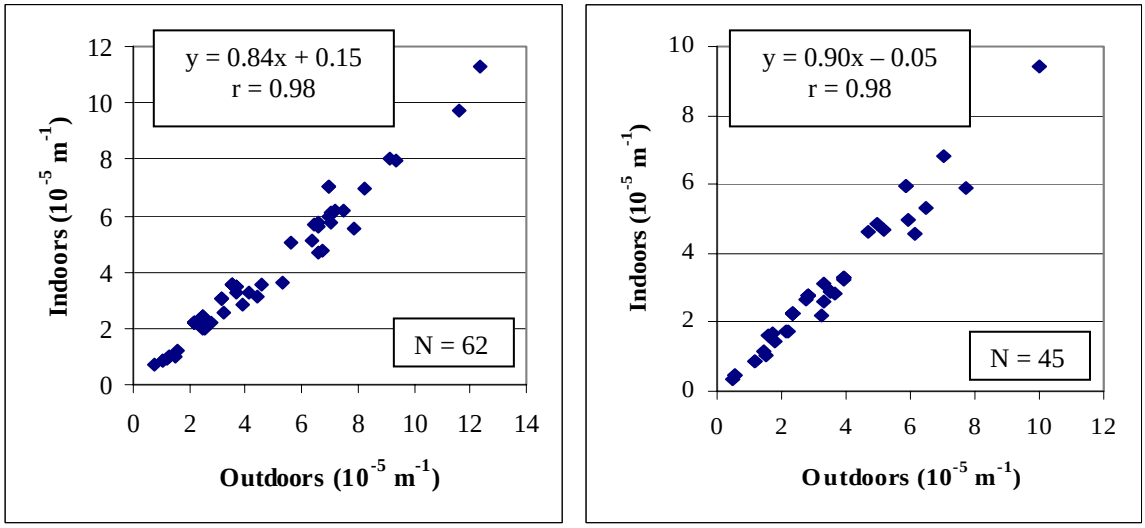
Regression analysis of all indoor and outdoor concentration data was conducted in order to further support the above-mentioned assumption. The corresponding regressions for PM mass and particulate EC (absorption coefficient values) are presented in Figures 1a, b and 2 a, b respectively.

In effect, the calculated correlation coefficients were significant. An excellent correlation was observed between indoor and outdoor absorption coefficient values (r = 0.98 for both size fractions). This finding, along with slope values close to 1.00 (equal to 0.84 for PM_{10} and 0.90 for $PM_{2.5}$) and low intercept values, clearly indicated that there was a large contribution of the ambient atmosphere to the indoor levels and that a large fraction of particulate EC of outdoor origin penetrated the indoor microenvironments (Halios *et al*., 2009; Na & Cocker III, 2005; Geller *et al*., 2002; Gotchi *et al*., 2002).



(a) (b)

Figure 1. Regression of indoor and outdoor concentrations for PM_{10} (a) and $PM_{2.5}$ (b).



(a) (b)

Figure 2. Regression of indoor and outdoor absorption coefficient values for PM_{10} (a) and $PM_{2.5}$ (b)

The corresponding correlation coefficients for PM_{10} and $PM_{2.5}$ concentration levels were calculated equal to 0.87 and 0.89 respectively, demonstrating again a significant contribution of the ambient atmosphere. The intercept values were again very low. The only important difference regarding the regression analysis results of absorbance and PM concentrations related to the slope values, which were much lower in the case of PM mass (0.56 for PM_{10} and 0.67 for $PM_{2.5}$). The significant impact of the ambient atmosphere to the residential concentration levels was also confirmed by the study of the indoor and outdoor concentrations diurnal cycles for CR, reported in a previous work (Diapouli *et al*., 2008).

The regression analysis results confirm the differences in the behaviour of particles when entering an indoor microenvironment, in relation to their size distribution and their chemical composition. Particulate elemental carbon is a chemically stable component, which is mainly found in particles smaller than 1 μm in aerodynamic diameter. It is therefore expected that it will penetrate indoors easier than larger particle fractions. Similarly, a higher fraction of ambient $PM_{2.5}$ may penetrate indoors in comparison to PM_{10}, which, due to their larger size, are obstructed when entering.

CONCLUSIONS

The aim of the present work was the characterization of PM (PM_{10} and $PM_{2.5}$) and elemental carbon (EC) indoor and outdoor concentration levels in the Athens area, at two critical for personal exposure microenvironments: the residential and office microenvironment. Two residences, one in a central area and one in a quite suburban area, as well as one office in the commercial center of Athens, were studied. Indoor microenvironments were selected as to present minimum indoor particle generation, in order to reveal the impact of ambient air to indoor concentration levels, through the examination of indoor / outdoor data relationships.

The results showed very high ambient levels at both central sites and significant levels at the suburban residential area, with the mean values in all three sites surpassing the E.U. 24-hr air quality standard for PM_{10} (50 μg m^{-3}). Exceedance of the 24-hr limit value was observed for 84 and 100 % of the measurement days at CR and CO respectively. Significant ambient concentrations were also recorded for $PM_{2.5}$ at both central sites (mean concentrations were greater than 25 μg m^{-3}, which is the E.U. annual limit value for this size fraction, to be achieved during 2010 – 2020).

The measured absorption coefficient values at the two central sites were also high, in comparison to relative results presented in the international literature, indicating a large contribution of elemental carbon in PM mass.

The measured indoor concentration levels were lower than the outdoor ones, as expected since there was minimum activity in the indoor microenvironments. Nevertheless, the results indicated significant population exposure indoors. High concentration levels of both size fractions were observed in the two central sites. Exceedance of the 24-hr limit value for ambient PM_{10} concentration was observed during 58 % and 23 % of the days, at CR and CO respectively.

Absorption coefficient values for the indoor filters were slightly lower than the respective outdoor values. The influence of traffic-related air pollution to indoor air quality was clearly reflected by the significant difference between central and suburban site results.

The value of the correlation coefficient between indoor and outdoor data may be used as an indicator of the influence of ambient air to the concentration levels measured in an indoor microenvironment, for different PM species. According to the regression analysis of indoor and outdoor PM and EC surrogate data, a large fraction of the ambient PM pollution penetrated indoors causing elevated indoor concentration levels. This phenomenon was more pronounced in fine particles, which have been also proved to have greater potency to cause adverse health effects.

In view of the special role of the indoor microenvironment to total personal exposure, a thorough investigation of the parameters influencing indoor concentration levels is needed. Recent epidemiological studies have indicated that particles of indoor and outdoor origin may differ in their biological effects, due to their different sources, size distribution and chemical composition. The examination of indoor and outdoor data relationships for different PM constituents and size fractions may give an insight into population exposure in indoor microenvironments and the special role of outdoor PM sources. The final aim is a better understanding of the indoor and outdoor generated particles behaviour, which is crucial for population risk assessment, since it may enhance the development of effective mitigation measures and the implementation of specific practices that will lead to an integrated policy for the protection of public health.

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