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Indoor and outdoor PM mass and number concentrations at schools in the Athens area

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Abstract Simultaneous indoor and outdoor PM_{10} and $PM_{2.5}$ concentration measurements were conducted in seven primary schools in the Athens area. Both gravimetric samplers and continuous monitors were used. Filters were subsequently analyzed for anion species. Moreover ultrafine particles number concentration was monitored continuously indoors and outdoors. Mean 8-hr PM_{10} concentration was measured equal to 229±182 μg/m^3 indoors and 166±133 μg/m^3 outdoors. The respective $PM_{2.5}$ concentrations were 82±56 μg/m^3 indoors and 56±26 μg/m^3 outdoors. Ultrafine particles 8-h mean number concentration was measured equal to 24,000±17,900 particles/cm^3 indoors and 32,000±14,200 particles/cm^3 outdoors. PM_{10} outdoor concentrations exhibited a greater spatial variability than the corresponding $PM_{2.5}$ ones. I/O ratios were close or above 1.00 for PM_{10} and $PM_{2.5}$ and smaller than 1.00 for ultrafine particles. Very high I/O ratios were observed when intense activities took place. The initial results of the chemical analysis showed that SO_4^{-2} accounts for the 6.6±3.5% of the PM_{10} and NO_3^1 for the 3.1±1.4%. The corresponding results for $PM_{2.5}$ are 12.0±7.7% for SO_4^{-2} and 3.1±1.9% for NO_3^-. $PM_{2.5}$ SO_4^{-2} indoor concentrations were highly correlated with outdoor ones and the regression line had the largest slope and a very low intercept, indicative of no indoor sources of fine particulate SO_4^{-2}. The results of the statistical analysis of indoor and outdoor concentration data support the use of SO_4^{-2} as a proper surrogate for indoor PM of outdoor origin.

Keywords Children exposure · Indoor and outdoor PM mass and number concentrations

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Introduction

There is an increased concern over the adverse health effects of air pollution, especially in urban areas, where many sources of air pollutants are concentrated. Suspended particles in particular have received much interest because of epidemiological and experimental evidence of their health impact. Both mass and number concentration of particulate matter (PM) have shown to correlate with acute health effects and measurable functional changes in the cardiovascular and respiratory system (Pope et al. 2002).

Studies conducted by Donaldson and McNee (1998) and Ferin et al. (1991) showed that, for the

same amount of PM mass deposited in the lung, toxicity tends to increase as particle size decreases. This may be attributed to the increased surface per unit mass (if the toxic components are located on the particle surface) or to the ability of finer particles to penetrate the lung tissues (Harrison and Yin 2000; Schwartz et al. 1996). In addition, numerous toxic and carcinogenic chemical compounds produced through combustion processes (e.g. toxic metals, PAHs) are mainly found to the submicron particle size fraction. Nevertheless, until now there are limited studies examining ultrafine particles (UFPs) concentrations levels, especially in indoor microenvironments. The first work that studied size distribution of particle number concentration and provided data on ultrafine particles number concentration levels in Greece was conducted by Grivas et al. (2004).

Children are one of the most sensitive population subgroups since they may receive an increased dose of PM to their lungs compared to adults. This may be due to greater fractional deposition with each breath and/or larger minute ventilation relative to lung size (Bennett and Zeman 1998). In effect, a 10-year longitudinal study in Southern California found deficits in growth of children's lung function associated with higher ambient concentrations of PM (Gauderman et al. 2000). Moreover, particulate air pollution has been found associated with increased respiratory symptoms, school absences and medication use for asthmatic children (Peters et al. 1997). Taking into account that a child spends most of its time indoors and especially in two microenvironments: the home and school-microenvironment, it becomes evident that the study of the indoor PM concentration levels must be a priority (Silvers et al. 1994; Farrow et al. 1997; Klepeis et al. 2001). On the other hand, in several studies conducted internationally, the PM concentrations measured inside classrooms were higher than the corresponding outdoor concentrations and concentrations measured inside residences, thus emphasizing the importance of the school-microenvironment in children's exposure (Roorda-Knape et al. 1998; Wheeler et al. 2000; Branis et al. 2005).

This work presents the first results of an ongoing study on schoolchildren exposure to PM in the area of Athens. The city of Athens has been facing air pollution episodes during the last three decades. Chaloulakou et al. (2003) conducted a one-year study of the PM_{10} and $PM_{2.5}$ concentration levels in downtown Athens. Both annual mean PM_{10} and $PM_{2.5}$ concentrations were found considerably higher than the corresponding European Union air quality annual standard for PM_{10} and the United States EPA annual $PM_{2.5}$ standard, respectively. Moreover, daily PM_{10} concentrations exceeded the E.U. limit for approximately 42% of the sampling days. The objectives of the present study are to characterize the PM_{10}, $PM_{2.5}$ and ultrafine particles concentration levels at elementary schools across the Athens area and to examine the relationship between the indoor and outdoor concentrations. The first results from the chemical analysis of the collected filters are also presented. Particulate sulfate concentration indoors and outdoors is of special interest since this chemical species is thought to be a good surrogate for indoor PM of outdoor origin (Suh et al. 1993; Long et al. 2001).

Materials and methods

Site description

Seven primary schools were chosen for measurement. An effort was made to include areas with different characteristics of urbanization and traffic density in order to obtain a general as possible picture of the exposure in the different areas of the Athens Basin. The characteristics of each site are described in Table 1.

Table 1 Characteristics of the different measurement sites

Site	Location	Description
RU	NORTH	Rural area, with no commercial activity and low traffic. There is unbuilt ground around the school
HI	WEST	Two blocks away from a major highway
RE	EAST	Residential area, surrounded by two major roads
UR	CENTER	Heavy-trafficked neighborhood in the center of Athens
MO	EAST	Residential area close to a major motorway
HP	SOUTH	Densely populated area close to a major motorway, with high commercial activity and heavy traffic
HA	SOUTH	At the harbor of Athens

Indoor samples were taken inside classrooms, at table height. In three schools, different rooms were selected for measurement in order to examine the indoor concentration differences in relation to the activities taking place inside. The selected microenvironments were a computer lab and a library, used by students only part of the school day, a teachers office and a gymnasium, where intense activity took place almost all day. No ventilation system existed in any of the schools. Outdoor measurements were carried out in the yard of each school, usually at an area not frequently visited by the students, for security purposes.

Sampling

The measurement campaign was conducted in two winter periods: November 2003–February 2004 and October–December 2004. Each school was studied for 2–5 consecutive weekdays during school hours (8:00 A.M. to 4:00 P.M.). Details on the measurement program (sampling period, number of sampling days, selected microenvironments, pollutants monitored) are presented in Table 2.

Simultaneous PM_{10} and $PM_{2.5}$ indoor and outdoor measurements were taken using Harvard PEMs at a flow rate of 4 l/min. The particles were collected onto 37 mm 2 μm pore size Teflon filters. All filters were pre- and post-conditioned in a clean room with environmentally controlled temperature and humidity prior to weighing. Weighing was carried out with an electronic microbalance with 0.01 mg resolution. Chemical analysis of the collected filters is under progress. Part of the filters is already analyzed for SO_4^{-2} and NO_3^- with the use of ion chromatography (IC). Details of the measurements can be found at Kouvarakis and Mihalopoulos (2002).

PM_{10} and $PM_{2.5}$ concentrations were also monitored continuously inside the schools and, in some of the schools, outdoors as well, by DustTrak (TSI, Model 8520). Similarly, ultrafine particles (0.01–1 μm) number concentration was measured continuously, by a Condensation Particle Counter (TSI, Model 3007), indoors at all schools and outdoors at only some of them. Both instruments were programmed to record every 1 min.

Results and discussion

Mass concentration

Mean 8-h PM_{10} concentration was measured equal to 229±182 μg/m^3 indoors and 166±133 μg/m^3 outdoors. The respective $PM_{2.5}$ concentrations were 82±56 μg/m^3 indoors and 56±26 μg/m^3 outdoors. Mean indoor and outdoor, PM_{10} and $PM_{2.5}$ concentrations for each school are presented in Figs. 1 and 2. PM_{10} outdoor concentrations were highly variable across the different school areas. The largest PM_{10} outdoor concentrations were observed at HP and MO, both areas affected by high traffic. As expected, $PM_{2.5}$ outdoor concentrations exhibited less variability across the different areas under study. Maximum PM_{10} mean indoor concentration was measured again at HP. Elevated indoor PM_{10} and $PM_{2.5}$ concentrations were also measured in a teachers office at RE, where smoking was allowed and at HI, where indoor measurements were conducted inside a gymnasium. It is worth noting the great effect of smoking on indoor $PM_{2.5}$ concentrations. The lowest indoor concentrations were noticed in a PC Lab at UR, which was occupied only part of the school day.

Table 2 Details on measurement program

Site	Number of sampling days	Indoor microenvironment	Monitored pollutants		Measurement
			Indoors	Outdoors	Period
RU	5	Classroom	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$	Dec. 2003–Jan. 2004
HI	5	Gymnasium	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$	Feb. 2004
RE	5	Library and Office	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$	Feb. 2004
UR	5	PC Lab	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$	Nov.–Dec. 2003
MO	2	Classroom	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$, UFPs	Jan.–Feb. 2004
HP	2	Classroom	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$, UFPs	Feb. 2004
HA	2	Classroom	PM_{10}, $PM_{2.5}$, UFPs	PM_{10}, $PM_{2.5}$, UFPs	Jan. 2004

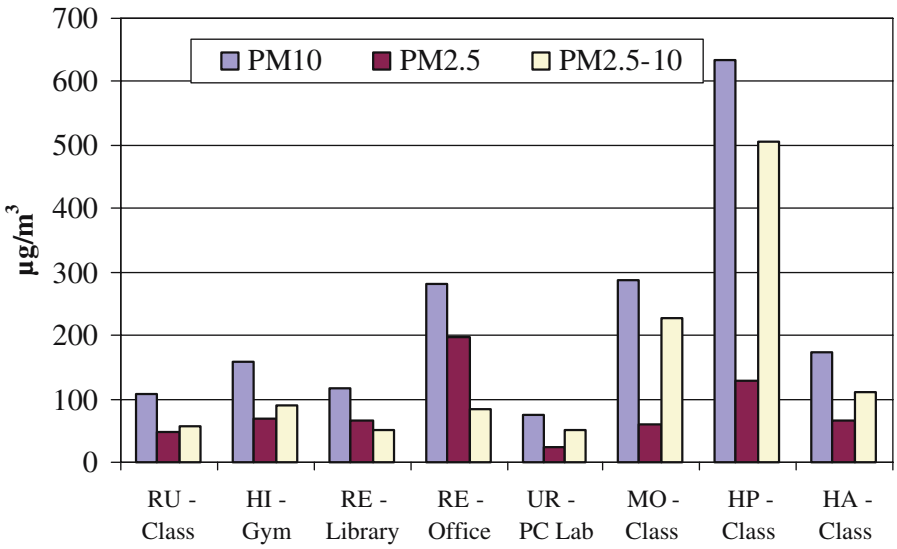

Fig. 1 Mean indoor PM_{10}, $PM_{2.5}$ and $PM_{2.5–10}$ concentrations

It is known that indoor PM levels are influenced by indoor particle generation and by infiltration of outdoor air. Particle generation indoors can be caused by specific sources (such as wood combustion and environmental tobacco smoke) or by human activities. It has been shown that in indoor microenvironments where there is no specific indoor source of pollution (e.g. smoking or combustion processes), people's activities may represent an important source of suspended particles (Monn et al. 1997; Luoma and Batterman 2001; Branis et al. 2005). Even the very presence of people in an indoor microenvironment may cause elevated PM concentration levels. Thus, the high indoor PM_{10} and $PM_{2.5}$ concentrations presented in the present work may be attributed to resuspension and generation of particles by the presence of students inside the classrooms.

The measured indoor and outdoor concentrations were significant for both PM_{10} and $PM_{2.5}$, when compared with results from similar studies conducted internationally. PM_{10} daily mean concentrations inside classrooms ranged from 21 to 617 $\mu g/m^3$ in Hong Kong (Lee and Chang 2000) and from 12.5 to 54 $\mu g/m^3$ in Detroit (Yip et al. 2004). Wheeler et al. (2000) measured in the U.K., during wintertime, mean PM_{10} and $PM_{2.5}$ classroom concentrations equal

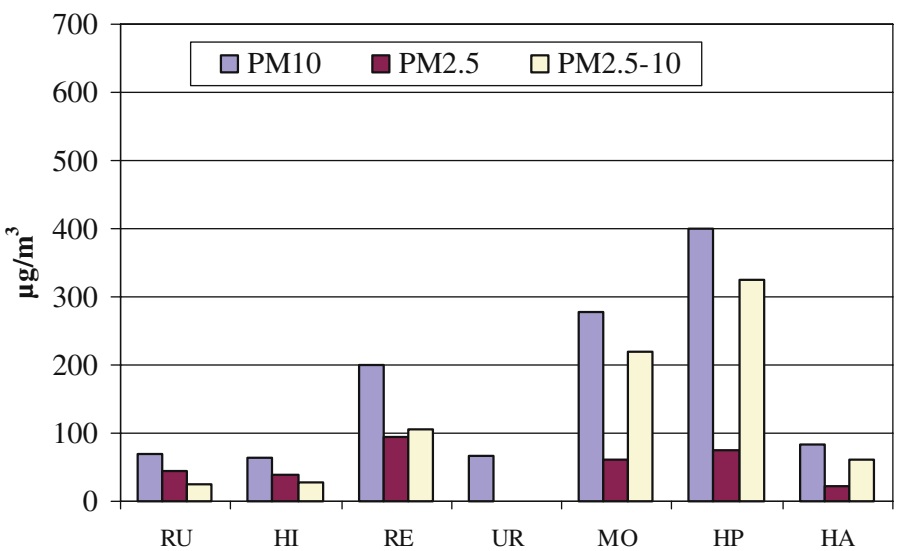

Fig. 2 Mean outdoor PM_{10}, $PM_{2.5}$ and $PM_{2.5–10}$ concentrations

Table 3 Indoor and outdoor concentration levels of PM_{10} and $PM_{2.5}$, SO_4^{-2} and NO_3^- ($\mu g/m^3$)

	Indoors	Outdoors
$PM_{10}SO_4^{-2}$	5.2±1.4	6.7±2.6
$PM_{2.5}SO_4^{-2}$	4.8±1.7	5.7±2.7
$PM_{10}NO_3^-$	2.3±1.8	3.8±2.2
$PM_{2.5}NO_3^-$	1.6±1.7	2.0±1.8

to 80 and 30 $\mu g/m^3$, respectively. Roorda-Knape et al. (1998) studied exposure to air pollutants near major motorways in The Netherlands. Mean PM_{10} concentrations at different schools ranged from 50.9 to 165.6 $\mu g/m^3$. In a more recent study, Janssen et al. (2001) studied $PM_{2.5}$ concentration levels at schools located close to motorways. The measured weekly indoor and outdoor concentrations were equal to 23± 6.3 $\mu g/m^3$ indoors and 24.8±11.6 $\mu g/m^3$ outdoors.

Chemical analysis of the obtained filters is under process. The initial results, from the chemical analysis of half of the filters collected at the first four schools (RU, HI, RE, UR), showed that SO_4^{-2} accounts for the 6.6±3.5% of the PM_{10} and NO_3^- for the 3.1±1.4%. The corresponding results for $PM_{2.5}$ are 12.0±7.7% for SO_4^{-2} and 3.1±1.9% for NO_3^-. PM_{10} and $PM_{2.5}$ SO_4^{-2} concentrations were equal to 6.1±1.4 $\mu g/m^3$ and 5.1±1.5 $\mu g/m^3$, respectively. Particulate NO_3^- was equal to 3.5±2.0 $\mu g/m^3$ in PM_{10} and 2.0±1.5 $\mu g/m^3$ in $PM_{2.5}$. SO_4^{-2} and NO_3^- concentration levels, indoors and outdoors, in PM_{10} and $PM_{2.5}$, are presented in Table 3. It is worth noting the high contribution of SO_4^{-2} to $PM_{2.5}$ (two times the corresponding contribution to PM_{10}). Studies conducted in PM collected at several outdoor locations in the Mediterranean (Mihalopoulos unpublished data) indicated that sulphate is the predominant form of sulphur. Thus the measured sulphate levels correspond to the PM total sulphur.

Number concentration

Ultrafine particles number concentration was measured equal to 24,000±17,900 particles/cm^3 indoors and 32,000±14,200 particles/cm^3 outdoors. Mean indoor and outdoor, ultrafine particles number concentrations for each school are presented in Fig. 3. The highest mean indoor ultrafine particles concentrations were observed in a small library room, with a carpet-covered floor and a very high ratio of persons per m^3 of space (51,900 particles/cm^3) and in the smoking office (52,090 particles/cm^3). The higher outdoor ultrafine particles concentration (42,700 particles/cm^3) was measured in HP, a heavy-trafficked and densely populated area. The measured average ultrafine particles concentrations were significant, especially when compared to results found in other international studies. Riesenfeld et al. (2000) studied indoor and outdoor ultrafine particles number concentration levels in Rochester, N.Y. The measured concentration inside a hospital, outdoors and inside a specially designed, very clean chamber were 3,600±

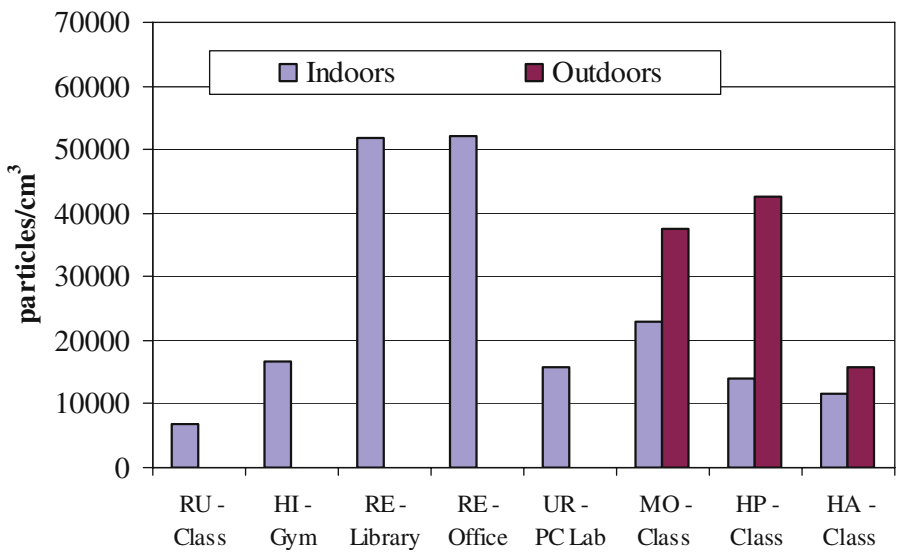

Fig. 3 Mean indoor and outdoor ultrafine particles number concentrations

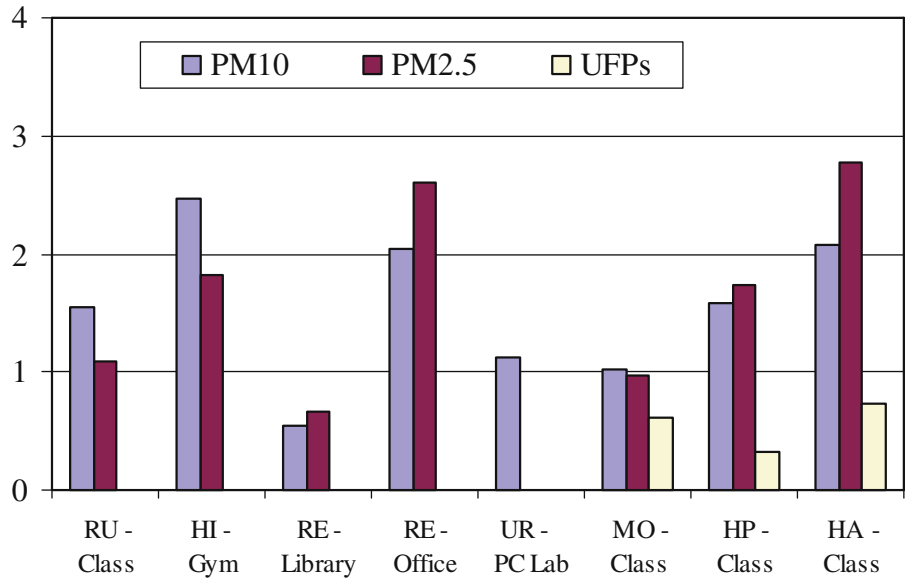

Fig. 4 Mean indoor-to-outdoor concentration ratios

2,700 particles/cm^3, 30,500±66,500 particles/cm^3 and 600±250 particles/cm^3, respectively. Ruuskanen et al. (2001) measured outdoor $PM_{2.5}$ mass and ultrafine number concentrations in three European cities. The $PM_{2.5}$ mean 24-hr concentrations were equal to 27.0±17.2 μg/m^3 in Alkmaar, 41.9±27.1 μg/m^3 in Erfurt and 9.4±5.0 μg/m^3 in Helsinki. The respective particle number concentrations were 25,800±11,300 particles/cm^3, 25,900±12,200 particles/cm^3 and 20,300±8,200 particles/cm^3.

Indoor to outdoor concentration ratios

Mean indoor-to-outdoor (I/O/) ratios varied depending on the indoor activities and the outdoor concentration levels. For PM_{10} I/O ratios ranged from 0.54 in RE (library microenvironment), where construction activities near the school resulted in elevated PM_{10} and $PM_{2.5}$ concentrations outdoors, to 2.46 in HI, where indoor measurements were conducted in a gymnasium with intense activity causing high indoor

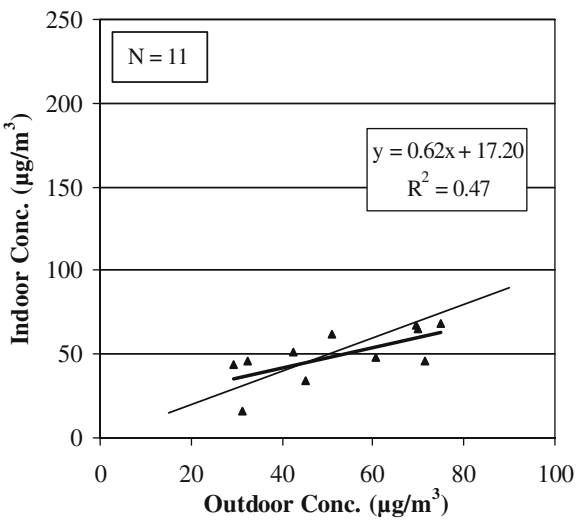

Fig. 5 Indoor versus outdoor concentration levels for $PM_{2.5}$ (1–1 line and regression line)

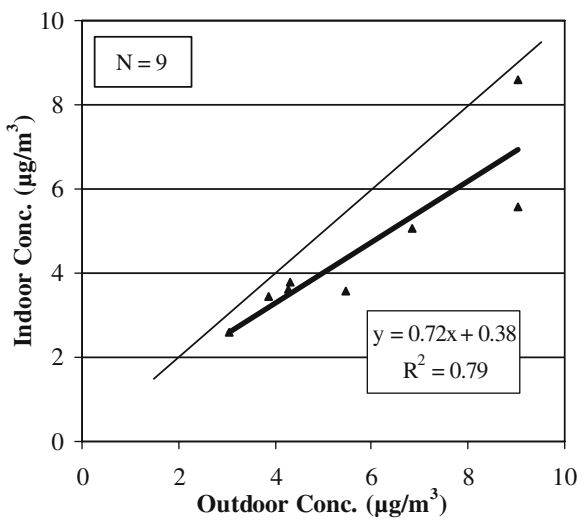

Fig. 6 Indoor versus outdoor concentration levels for SO_4^{-2} in $PM_{2.5}$ (1–1 line and regression line)

PM concentrations. The respective I/O ratios for $PM_{2.5}$ varied from 0.67 to 2.77, with the lowest value corresponding again to the library at RE. The highest I/O ratios were observed again at RE, in the smoking teachers office (2.60) and at HA (2.77), where outdoor $PM_{2.5}$ concentrations were relatively low. I/O ratios for ultrafine particles ranged from 0.33 to 0.74. The high I/O ratios for both PM_{10} and $PM_{2.5}$ are due to the presence of children and the activities taking place inside classes. On the other hand, ultrafine particles' main source is presumed to be vehicular traffic (Fig. 4).

Correlations between indoor and outdoor concentrations for PM_{10} and $PM_{2.5}$ were also examined. As it was expected, the slope of the PM_{10} regression line was smaller than the one of $PM_{2.5}$. Indoor versus outdoor concentration levels for $PM_{2.5}$ and SO_4^{-2} in $PM_{2.5}$ are presented in Figs. 5 and 6, respectively (The dark line corresponds to the regression equation, while the gray line is the 1–1 line). The two regressions are statistically significant ($p<0.001$). The data used in these two figures are 8-h mean daily concentrations and refer to the days when analysis on the filter SO_4^{-2} concentrations was conducted. $PM_{2.5}$ SO_4^{-2} indoor concentrations were highly correlated with outdoor ones and produced the largest slope and a very low intercept (7% of the mean outdoor SO_4^{-2}), which is indicative of no indoor sources of fine particulate SO_4^{-2}. Therefore, all indoor $PM_{2.5}$ SO_4^{-2} concentration could be considered of outdoor origin. This result is in good agreement with studies proposing fine particulate sulfate as a surrogate for indoor PM of outdoor origin (Suh et al. 1993; Long et al. 2001; Sarnat et al. 2002; Hanninen et al. 2004; Sawant et al. 2004).

Conclusions

The present work examines PM_{10}, $PM_{2.5}$ and ultrafine particles concentration levels at seven schools located in areas with different degrees of urbanization and traffic density, to characterize the children exposure levels in the area of Athens. High levels of PM_{10}, $PM_{2.5}$ and ultrafine particles concentrations were measured both indoors and outdoors. As expected, outdoor PM_{10} concentrations exhibited a greater spatial variability than the corresponding $PM_{2.5}$ ones. The indoor PM concentration levels varied depending on the indoor activities and the outdoor concentrations. The higher PM_{10} indoor and outdoor concentrations were measured at HP, a densely populated area close to a major motorway, with high commercial activity. HP presented also the higher outdoor ultrafine particles concentration, while the higher indoor ultrafine particles concentrations were observed in a small library room, with a carpet-covered floor and a very high ratio of persons per m^3 of space.

I/O ratios for PM_{10} and $PM_{2.5}$ were close or above 1.00 at almost all sites. The higher I/O ratios were found in the gymnasium (2.46), where there was intense activity, leading to elevated indoor concentrations, in a smoking office (2.60) and at HA (2.77), where the outdoor concentrations were low. The corresponding ratio for ultrafine particles was smaller than 1.00 in all cases, since vehicular traffic is presumed to be UFPs main source.

SO_4^{-2}% contribution to $PM_{2.5}$ mass was almost two times the respective one for PM_{10}. SO_4^{-2} indoor concentrations were strongly associated with outdoor ones and produced the largest slope and a very low intercept. All these results support the use of SO_4^{-2} as a proper surrogate for indoor PM of outdoor origin. However, due to the limited available data used in the statistical analysis, these results must be considered with caution.

This work is the first effort to study schoolchildren exposure to PM in Athens. The high levels of PM_{10} and $PM_{2.5}$ concentrations, both indoors and outdoors of attended schools, rises a great concern and needs the attention of public health authorities. The protection of indoor air quality in a microenvironment so critical and characteristic for children total daily exposure, must be a first priority goal and demands the design and implementation of intervention actions in the near future.

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