Outdoor/indoor relationships of PM_1 and black carbon: sources and infiltration processes in an urban environment

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Abstract

Ambient air emissions of sub-micron particles infiltrate into indoor environments and play a major role in indoor air quality. Discriminating between particles of indoor *vs.* outdoor origin is therefore essential when assessing indoor air pollutant levels and characteristics. Outdoor/indoor relationships of traffic-derived nanoscaled particulates (PM_1 and black carbon) were determined in a typical Mediterranean environment. Results evidenced the major impact of outdoor vehicular traffic emissions on indoor particulates: despite windows remaining closed at all times, 70% of indoor BC and 73% of indoor PM_1 originated from outdoor emissions. This was probably due to due to inadequate insulation of the building. Outdoor/indoor penetration ratios were relatively constant for BC (1.29±0.08) but not for PM_1 (1.95±0.38), suggesting it is advisable to monitor the variability of penetration factors over time. Particle infiltration seemed to depend not only on physical barriers (building envelope, ventilation systems, etc.), but also on the physico-chemical properties of the particulates. Printing and photocopying contributed with 25-30% (546 ng/m^3) of total indoor BC. Dust re-suspension by worker passage was the main indoor source of PM_1 (15-20%, 1.1 µg/m^3).

Keywords: exhaust, infiltration, penetration ratio, Barcelona, Spain, shift analysis.

Introduction

Numerous research works available in the literature have reported significant health risks associated with exposure to particulate matter (PM) (Brunekreef et al., 1997; Pekkanen 1997; Lippmann & Schlesinger, 2000; Künzli et al., 2000; Pope & Dockery, 2006). In recent years research interest has been directed towards fine and ultrafine particles (particulates with aerodynamic diameter <1µm and <0.1µm, respectively). Epidemiological studies (Donaldson et al., 2001; Oberdoster, 2001) indicate that fine and ultrafine particles may have a greater potency to cause adverse health effects than larger particles, due to their increased deposition rates in the lower respiratory tract (Jaques & Kim, 2000) and their higher transition metal content (Shi et al., 2004). Furthermore, ultrafine particles (UFP) have also been suggested to be more toxic because of the large surface area available for biologic interactions with lung cells (Chio et al., 2008). There is little information on the trend in UFP in European urban atmospheres, but the increased load of diesel vehicles and recent data (Mejia et al., 2007) suggest an upward trend. Recent works have shown that traffic is the origin of 90% of the UFP load in urban environments (Pey et al., 2009).

Population exposure to both supra-micron and nanoscaled particulate concentrations is usually considered to take place in outdoor environments. However, it is estimated that on average adults spend 60-90% of their time indoors (Klepeis et al., 2001). Whereas data on outdoor fine and ultrafine particle levels and physico-chemical characteristics are widely available in Europe (Putaud et al., 2004; Querol et al., 2004; Hueglin et al., 2005; Andersen et al., 2007; Lonati et al., 2007), fewer data are available in the literature regarding the levels and characteristics of atmospheric particulates in indoor air However, the interest on this topic is increasing in the US (Jones et al., 2000; Geller et al., 2002; Sawant et al., 2004; Polidori et al., 2006; Lunden et al., 2008; Martuzevicius et al., 2008), Europe (Fischer et al., 2000; Kingham et al., 2000; Cyrys et al., 2004; Hänninen et al., 2004; Hoek et al., 2008) and Asia (Chao & Tung, 2001; Cao et al., 2005; Chen & Zhao, 2011; Chen et al., 2011). The lower number of studies focusing on indoor in comparison to outdoor air is generally attributed to the complexity in the determination of PM levels indoors, given the broad variability of direct indoor sources (printer emissions, dust re-suspension, smoking, cleaning, etc.) as well as the multiplicity of microenvironments (office spaces, industrial facilities, households, etc.). Evidently, particle levels in indoor environments are greatly influenced by indoor activities, which are characterised by typically generating high-concentration but short-term pollution events (acute events; Long et al., 2000). The examination of short-term variations in particulate concentrations is, therefore, essential in indoor air quality studies. In addition, indoor levels of UFPs may also be greatly influenced by outdoor sources as a function of building and ventilation characteristics and habits (Morawska et al., 2001; Lunden et al., 2008; Martuzevicius et al., 2008; McNabola et al., 2011), by three major mechanisms: mechanical ventilation, natural ventilation, and infiltration. Three parameters are generally used to monitor the presence of outdoor particles in the indoor environment: the indoor/outdoor (I/O) ratio, the penetration factor and the infiltration factor (Chen & Zhao, 2011). The major outdoor emission source of fine and ultrafine particles is vehicular traffic, which is considered a global public health problem (World Bank 2006) and one of the major sources of particulate matter in urban areas worldwide, and in particular in Barcelona (Amato et al., 2009).

In this respect, the influence of outdoor PM sources on indoor environments is a key issue to be taken into consideration. Over the past several decades, the influence of outdoor air pollutants in indoor environments is believed to have decreased due to a variety of factors (Heinrich, 2010), including the construction of more tightly sealed buildings and reduced ventilation rates to save energy. However, Morawaska et al. (2001) studied the indoor/outdoor (I/O) relationships in the residential environment in Australia, and found ratios varying from 0.2 to 2.5 and depending on particle size distribution. Other studies have been conducted (Quackenboss et al., 1989; Wallace, 1996), which indicate that the ratio of the indoor to outdoor particle mass concentrations varies in a wide range from 0.5 to 2 in the absence of indoor particle sources. Conversely, in the presence of indoor sources such as smoking or other indoor combustion sources (e.g., fireplaces), I/O ratios may be as high as 31 (Chen & Zhao, 2011). More recently, I/O relationships have been studied in the US and Australia with a special focus on the penetration rates of traffic-related particulates (Geller et al., 2002; Baxter et al., 2008; Lunden et al., 2008; Martuzevicius et al., 2008; Morawska et al., 2009). In Asia, I/O ratios were found to vary considerably due to the difference in size-dependent indoor particle emission rates, the geometry of the cracks in building envelopes, and the air exchange rates, thus making it difficult to understand the indoor/outdoor relationship using the I/O ratio only (Chen & Zhao, 2011). As for Europe, fewer studies are available and they focus mainly on modelling indoor air quality in indoor environments (Diapouli et al., 2007; Bohlin et al., 2008; Orosa & Baaliña, 2008), and on indoor air quality assessments in schools (Poupard et al., 2005; Fromme et al., 2007; Almeida et al., 2010). Studies on indoor air quality in workplace environments are especially rare in Southern-European countries, which are of special interest due to the influence of the Mediterranean climate on building ventilation systems (frequent air conditioning and natural ventilation by open windows).

While most of the available studies focus on particle mass, there is still very little information on the indoor/outdoor relationship for other parameters such as particle composition, emission sources, particle number or surface area. However, UFPs are enriched in organic carbon content as well as prooxidative polycyclic aromatic hydrocarbons (PAH) that promote oxidative stress and inflammation, suggesting that it is not only the particle mass but also particle composition (in particular, carbon content) which should be considered when assessing indoor air quality and potential health effects. Understanding the relationship between outdoor and indoor aerosol particles for parameters other than PM mass is of importance to develop efficient control strategies to reduce health risk.

Based on the above considerations, the main objective of the present work is to understand the role of outdoor and indoor emission sources as determinants of indoor air quality in an urban environment. Specifically, this work focuses on the identification and characterisation of outdoor and indoor sources of fine and ultrafine particles in an indoor environment (a workplace in a typical Mediterranean urban area), and on understanding the factors influencing particle infiltration from ambient air into indoor environments. Going beyond the state-of-the-art, indoor air quality was assessed in terms of nanoscaled particulate mass (<1 micron, PM_1) and black carbon (BC) concentrations.

Materials and methods

Simultaneous monitoring of outdoor and indoor PM_1 and BC concentrations was carried out at a chemical research facility (IDAEA-CSIC), in the urban area of Barcelona (NE Spain). The building dates back to 1967, and has undergone no renovations ever since. The indoor location was a vacant laboratory located on the 5th floor, adjacent to other laboratories and offices which were in use on a daily basis during the study period. The studied laboratory (vacant) had the same dimensions, geometry, windows and doors as the laboratories in use, and was connected to them by means of a corridor. The doors of the vacant laboratory were kept open at all times in order to ensure air mass circulation with the remaining offices and laboratories on the same floor (which were occupied at the time). Windows were kept closed at all times, and there was no air conditioning. Indoor BC and PM_1 concentrations were compared with outdoor BC and PM_1 levels from an urban background monitoring station located at <150m from the research facility. The background station was located in a park in the vicinity of one of the major traffic arteries in the city, with a mean flow of approximately 100.000 vehicles/day.

Two sets of identical instruments were deployed at the indoor and outdoor monitoring stations. Black carbon concentrations were monitored using two multi-angle absorption photometers (MAAP) with a 10-minute (indoor) and 1-hour (outdoor) time resolution. The noise of the MAAP has been determined to be 0.22 Mm^{-1} for 1 min averaging interval (Müller et al., 2010), and the uncertainty of the instrument is ±12% (Schladitz et al., 2009). PM_1 concentrations were monitored by means of two laser spectrometers (GRIMM 1107) with 15-minute (indoor) and 30-minute (outdoor) time resolutions. The laser spectrometers are based on single particle detection and counting system by laser light scattering at 90º, which classifies the particles by size distribution into 31 channels and generates as output PM_1, $PM_{2.5}$ and PM_{10} mass concentrations based on the corresponding specific density factors.

The measured particle size range is 0.25 to >32 µm (according to the manufacturer), with a sampling efficiency >95% (Bukowiecki et al., 2011). All instruments were serviced before the monitoring campaign. Both laser spectrometers are routinely calibrated by comparison with reference gravimetric high-volume samplers, and corrected by means of a factor. For the purpose of this study, one high-volume sampler was set up in the indoor environment in order to obtain the indoor-specific correction factor. The temporal resolution of the instruments located indoor was set higher than outdoor aiming to observe short-term emissions generated by indoor sources. Monitoring of BC and PM_1 was carried out during a 4-month period between 22/03/2010 and 28/07/2010 (with breaks due to technical issues).

In addition to online monitoring of BC and PM_1, $PM_{2.5}$ aerosols were collected on filter substrates for chemical analysis. In the indoor environment, one low-volume $PM_{2.5}$ sampler (1 m^3/h) collected samples on 47-mm quartz fibre filter substrates during the day (9:00-17:00h, local time) and night (18:00-8:00h) shifts. In total, 66 valid samples were collected. In the outdoor site, $PM_{2.5}$ samples were collected over 24-hr periods by means of a high-volume sampler (MCV SA, 30 m^3/h). A total of 30 valid samples was collected. The $PM_{2.5}$ mass was determined by gravimetry after the filters (samples and blanks) had acclimatised at controlled temperature conditions for at least 24h. The chemical composition of the particulates collected on the filters (indoor and outdoor) was characterised by means of inductively-coupled plasma mass and atomic emission spectrometry (ICP-MS and ICP-AES, for major and trace elements), and ion chromatography (IC) and flow injection analysis (FIA) for water-soluble ions, according to the methodology described in Querol et al (2001). A detailed description of the uncertainties associated with the PM mass and chemical analyses is provided elsewhere (Viana et al., 2006; Minguillón et al., 2007). Finally, 8 $PM_{2.5}$ 24-hr samples were collected in the indoor environment by means of a reference high-volume sampler. The aim was to compare the online $PM_{2.5}$ data and the low-volume mass concentrations with those obtained with the reference high-volume sampler, and thus obtain correction factors for the non-reference instruments.

Results and Discussion

Indoor and outdoor BC and PM_1 concentrations

Mean, minimum and maximum levels of BC and PM_1 at the indoor and outdoor locations are summarised in Table 1. Indoor and outdoor PM_1 levels, determined by laser spectrometry, were corrected with respect to the reference gravimetric method by applying the same correction factor obtained for $PM_{2.5}$ (indoor r^2=0.86; y=0.67x+2.24; outdoor r^2=0.77; y=0.88x-0.63, Figure 1).

On average, BC and PM_1 levels were higher in the outdoor environment than indoors (19% and 43% higher, respectively), with mean levels for the entire monitoring period of 1572 ngBC/m^3 and 7.6 µgPM_1/m^3 indoors, and 1871 ngBC/m^3 and 12.5 µgPM_1/m^3 outdoors. Maximum BC concentrations (15- or 30-minute means) were, however, higher in the indoor environment (7% higher), as a result of the peak events described above. Mean indoor/outdoor (I/O) ratios for the entire monitoring period (calculated on a sample-by-sample basis) were 0.97 and 0.76 for BC and PM_1, respectively, which are higher than values found in the literature for European urban environments for BC (0.60-0.65, Lunden et al., 2008), and also higher than average for PM_1 but within the ranges described in the literature (0.14-1.37 for particles with aerodynamic diameter between 0.5 and 1.5 µm, Chen & Zhao, 2011). Out of the 17 studies reviewed in the work by Chen & Zhao (2011) on I/O ratios for particles between 0.5µm and 1.5µm, 60% (10 studies) reported I/O ratios <0.75. Thus, the higher I/O ratios obtained for PM_1 and BC in the present study evidence a major influence from outdoor particles in the indoor environment (especially BC), probably as a result of inadequate insulation of the building due to its old age (>40 years). Structural characteristics of the buildings such as insulation materials or ventilation patterns have been shown to play a more important role in affecting the indoor concentrations of the traffic-related aerosol, than parameters such as the distance to major roads or traffic density (Lunden et al., 2008; Martuzevicius et al., 2008). The ratios were calculated on a sample-by-sample basis, and then averaged (not calculated as the ratio of the means).

The time series of simultaneous indoor and outdoor PM_1 and BC concentrations were analysed in search for emission patterns. As shown in Figure 2, the correlation between indoor and outdoor measurements was significantly high, with good correspondence in absolute values (y=1.07x) although with a relatively high dispersion (r^2=0.35) in the case of BC. The results evidenced a clear daily pattern coinciding with vehicular traffic emissions, with maximum levels during traffic rush hours, especially visible for BC. The strong correlation between the indoor and outdoor levels of both parameters, as well as the similarity in the absolute concentrations, suggests vehicular traffic as the dominant and common emission source in both environments.

Infiltration of outdoor emissions

In order to quantify the influence indoors of outdoor PM_1 and BC emissions, penetration factors were calculated (Kuhlbusch et al., 2009). The aim was to determine the infiltration rate of outdoor particles into the indoor environment, in order to be able to: a) discriminate between the indoor particle emissions and the background levels of outdoor origin, and b) quantify the contributions from each of these sources in terms of PM_1 and BC. Outdoor-to-indoor ratios (O/I) were calculated for time periods during which no indoor PM emissions were generated, typically weekends or Sundays in industrial facilities and workplaces. In the facility under study, weekends were representative of periods with zero indoor-generated emissions. The O/I ratio for BC (Table 2, calculated for the 3 weekends for which simultaneous indoor and outdoor BC data were available) showed limited variation between May and July (spring *vs.* summer), with a mean value of 1.29 ± 0.08 (O/I = 1.20 to 1.35). Thus, the infiltration of BC particles was relatively constant and showed no seasonal dependence. Conversely, the O/I ratios for PM_1 (Table 2, 7 weekends available) showed a statistically significant decrease of 42% from April to July at a 0.01 confidence level according to the Mann Kendall test (Salmi et al., 2002). Whereas the mean O/I ratio for PM_1 was 2.13 in April, it was closer to 1 (1.14-1.23) in July, indicating a much higher degree of particle infiltration during summer than in spring. Given that windows remained closed at all times, this increase in particle infiltration was interpreted as a change in particle characteristics, either particle composition and/or size.

One possible interpretation for the different trends detected for the O/I ratios for BC and PM_1 could be the faster summer oxidation rates of SO_2 due to enhanced photochemistry, which results in larger contributions to PM_1 from sulphate particles with a finer grain size distribution and thus an enhanced infiltration from outdoors to the indoor environment. This process would not influence the penetration ratio of BC particles, with a grain size distribution which is unaffected by seasonal trends. In order to test this hypothesis, the sulphate and nitrate content of the indoor $PM_{2.5}$ aerosols was analysed (Figure 3), assuming a similar distribution in PM_1 based on the fine grain size distribution of atmospheric sulphate (Seinfeld & Pandis, 1998). Results showed a marked increase in the sulphate content of outdoor $PM_{2.5}$ aerosols, which accounted for 8% of $PM_{2.5}$ in March and increased progressively to 29% in July. At the same time, ambient levels of nitrate decreased from 14% (March) to 2% (July) in the $PM_{2.5}$ fraction as a result of the thermal instability of this species. In parallel to the trends observed in the outdoor environment, sulphate levels in indoor $PM_{2.5}$ (and thus in PM_1) increased between March (10% of $PM_{2.5}$) and June (22%), while the contribution from coarser nitrate particles decreased (from 19% to 4%). Thus, results evidenced the seasonal variability of the chemical composition of outdoor particulates, which implied a shift to a finer grain size distribution and thus resulted in higher infiltration rates into the indoor environment. Consequently, the chemical composition of indoor particles was also seasonally dependent.

In addition to this, it is worthy to note that the indoor-outdoor temperature differences cause two-way air flows and thus lead to a significant influence on particle and pollutant penetration or infiltration (Chen et al., 2011a,b). Consequently, the seasonality of the indoor-outdoor temperature differences should also affect the infiltration of outdoor particles in the indoor environment. In the present work air exchange ratios were not measured due to the fact that there was no direct mechanical or manual ventilation in the study laboratory, given that one of the main goals of the study was to assess the impact of the building envelope on indoor particle levels.

In conclusion, particle infiltration does not only depend on physical barriers (e.g., windows, insulation) determining the outdoor-indoor exchange of particulates, but also on the physico-chemical properties of those particles. Therefore, in order to accurately discriminate the contribution to indoor air quality of particulates originating from indoor and outdoor emissions, it is necessary to monitor the variability of the calculated penetration factors as a function of time. This is not the case for studies in industrial facilities, e.g., aiming to detect leaks in manufacturing processes, for which point measurements of penetration ratios (e.g., one weekend) are recommended (Kuhlbusch et al., 2009). Our results evidence that, when infiltration of outdoor particulates is a major source of indoor aerosols, the variability of penetration factors should be monitored over time.

Indoor BC and PM_1 emissions were estimated based on the penetration factors described above and the outdoor and indoor BC and PM_1 levels. Figure 4 shows an example of the indoor BC concentrations monitored (total), as well as the calculated indoor BC of outdoor origin and indoor BC of indoor origin, for the month of July (when simultaneous indoor and outdoor BC data were available). Results showed that, on average and for the entire monitoring period, 70% of the indoor BC was originated outdoors, and 30% was generated by indoor emissions. The influence of outdoor emissions was only slightly higher for PM_1, for which 73% of the indoor mass originated from outdoor sources. These values are much higher than those reported by other authors (34% of indoor $PM_{2.5}$ originating from outdoor traffic-derived sources, Martuzevicius et al., 2008). Even after the subtraction of the contribution from outdoor sources, indoor BC still showed peaks coinciding with vehicular traffic rush hours (Figures 4 and 5). This is a consequence of the application of mean weekend O/I ratios to hourly weekday data. Despite this artefact, this analysis allowed for the identification of indoor sources, as will be discussed below.

Indoor BC and PM_1 emission sources

The daily cycle of total indoor BC (Figure 5, daily mean=1572 ng/m^3) showed a clear traffic-derived trend, which contrasted with indoor BC emissions (daily mean=449 ng/m^3) which were characterised by one major increase between 6:00-12:00 UTC (8:00-14:00 local time) and lower levels in the afternoon and evening hours. The morning increase was interpreted as the reflection of workplace activities linked to BC sources (printing, photocopying, etc), which were not located directly in the study laboratory and therefore their contributions to BC levels were not observed as the sharp, short-term BC peaks characteristic of printer emissions (He et al., 2010). Most probably, printing and copying activities were carried out during the morning, generating the BC increase shown in Figure 5. These activities were not dominant after the lunch break (13:00-14:00 local time). Other potential BC sources may be present but were not identified by this analysis. The potential contamination due to the monitoring instruments (pumps) was estimated in a maximum constant contribution of 250 ng/m^3 (Figure 4). As described above, the influence of outdoor traffic-derived aerosols was still detected as a secondary BC peak at 6:00-7:00 UTC (8:00-9:00 local time). This indicates that particles infiltrated from the outdoor environment cannot be fully segregated by applying O/I ratios in this type of work environment, even though this methodology is successfully applied in industrial workplaces. The reason for this is that particles are emitted at much higher concentrations in industrial environments, and they dominate over particulates infiltrated from ambient air.

Contrarily to the BC daily cycle, total indoor PM_1 levels showed relatively small hourly variations (mean daily PM_1 = 7.3 µg/m^3, standard deviation = 0.32 µg/m^3), which result from the sum of the indoor and outdoor daily emission trends. In ambient air, PM_1 levels decreased between 10:00 and 19:00 UTC (12:00-21:00 local time) mainly as a result of dilution due to the increased boundary layer height at midday and in the afternoon hours. As for the indoor PM_1 emissions (daily mean=1.8 µg/m^3), their daily cycle was affected by the same artefact described for BC, and showed a slight impact of outdoor emissions at 7:00 UTC. Aside from this, PM_1 emissions peaked in parallel with those of BC between 8:00 and 12:00 UTC, probably as a result of worker passage along the corridor and between laboratories. Contrarily to BC, indoor PM_1 emissions remained relatively high (2-2.5 µg/m^3) in the afternoon hours, only to decrease at the end of the shift (17:00 UTC). Consequently, indoor air quality was affected by BC and PM_1 levels during the morning hours (until the lunch break), whereas in the afternoon hours and until the end of the shift indoor emissions were dominated by PM_1 sources.

Work day vs. weekend analysis

Due to the artefact described above, when the daily cycle of indoor BC was analysed separately for work days and weekends (Figure 6), the morning traffic peak was detected throughout the entire week (even though with different intensities) given that traffic emissions decrease on weekends, but do not cease. As expected, the traffic peak was slightly delayed (1 hour) on weekends, with respect to work days. Conversely, the 8:00-12:00 UTC peak (due to workplace activities) was only present during weekdays, being absent on weekends when the building was empty. In terms of mass, during the 8:00-12:00 period, BC levels were on average 546 ng/m^3 higher on work days with respect to weekends (considered the background levels) due to workplace activities (61% increase, on average 346 ng/m^3 on weekends and 892 ng/m^3 on work days). This increase accounted for 25-30% of the total indoor BC concentrations during the maximum indoor emission period (8:00-12:00 UTC). Similar results were obtained for PM_1, even though two unexplained indoor emission peaks were detected at 10:00-11:00 UTC and 15:00-16:00 UTC. Because of the extremely low variability of the levels of indoor PM_1 described in the previous section and Figure 5, these peaks could result from the uncertainty of the measurements. A rough estimation suggested that the increase of the indoor PM_1 levels as a result of indoor emissions could be 1.1 µg/m^3 (hourly mean, 15-20% of total indoor PM_1 during the maximum emission period). For BC, the results from this analysis confirm the interpretation of the indoor emission sources described in the previous sections, pointing towards printing and copying tasks as the major sources of indoor BC emissions.

Shift analysis

Epidemiological studies (Tovalin-Ahumada & Whitehead, 2007; Zhang et al., 2009) evidence the relevance taking into account the duration and time of work-shifts when assessing indoor air quality levels and their potential impact on human health. This type of analysis is especially relevant in industrial facilities, in which production is independent of the time of day. In the present study, a separate analysis of indoor air pollutants was carried out for the day (6:00-17:00 UTC) and night (18:00-05:00 UTC) shifts. During the night shift only the cleaning crew was present inside the building. Given that outdoor vehicular traffic was the main source of indoor BC (accounting on average for 70% of indoor BC), results showed that the levels of this pollutant in the indoor environment were the lowest during the night shift (on average 31% lower, 2008 ng/m^3 during the day shift and 1385 ng/m^3 during the night, Table 3). Because of the low hourly variability of indoor PM_1 levels shown in Figure 4, only minimal differences were observed between the day and night shifts (5%, 7.4 µg/m^3 *vs.* 7.0 µg/m^3, respectively). In conclusion, BC levels were lower during the night than during the day shift, resulting from the combination of minimal indoor emissions and reduced ambient levels (and thus, low infiltration of BC particulates). PM_1 levels were rather constant mainly due to the relative stability of indoor PM_1 concentrations. As for PM_1 constituents, the mean indoor PM_1 chemical composition was dominated by mineral matter (7.2 µg/m^3), probably as a result of dust re-suspension by worker passage. It is important to note that filter samples were only collected on work days (no weekend sampling), and therefore the PM_1 concentrations during the days with filter samples were higher than the average for the entire monitoring period (14.9 µg/m^3 for days with filter samples *vs.* 7.6 for the entire monitoring period, Table 1). On average, the contribution from secondary inorganic (sum of sulphate, nitrate and ammonium) and sea-salt aerosols to indoor air was similar in terms of mass (4.0 and 4.2 µg/m^3, respectively). When comparing day and night shifts, a marked decrease was observed for the mineral matter content (9.4 µg/m^3 during the day to 5.8 µg/m^3 during the night), thus confirming that dust re-suspension by the passage of workers along the corridor and within the laboratories is the main source of indoor PM_1. The sum of Na and Cl^- (initially grouped as marine aerosol) also showed a clear decrease (6.1 to 3.7 µg/m^3), indicating that the source of these components is not natural sea-salt only, and that significant contributions to the levels of Cl^- are derived from indoor emissions (given that the study location is a chemical research facility). Finally, the levels of secondary inorganic aerosols remained relatively constant between day and night (4.2-4.9 µg/m^3), given that the main source of these components in the indoor environment is infiltration from outdoor air.

Conclusions

The sources and infiltration processes of nanoscaled particulates (BC and PM_1) were determined in an indoor environment. A major impact of outdoor vehicular traffic emissions on indoor particulates was detected, which resulted from the inadequate insulation of the building. Even ensuring that windows were closed at all times, at least 70% of indoor BC and 73% of PM_1 concentrations originated from outdoor emissions. Outdoor to indoor infiltration of fine and ultrafine particles was higher for BC than for PM_1 (O/I = 1.29±0.08 vs. 1.95±0.38, calculated for “zero indoor emission” periods), probably due to its finer grain size distribution.

Particle infiltration showed no variation with time in the case of BC, but it showed a statistically significant decrease for PM_1 (O/I=2.13 to 1.23) from spring to summer. This was due to an increase in the contribution from fine grain-sized sulphate particles. We conclude that particle infiltration does not only depend on physical constraints but also on the physico-chemical properties of the particles.

The indoor emission sources of BC and PM_1 aerosols were also investigated. Printing and copying activities were identified as the main BC emission sources, and during their maximum emission period (10:00-14:00h) mean hourly BC levels increased by 71% with respect to the same period on weekends. Sources of PM_1 were mainly related to dust re-suspension by the passage of workers, occurring in the morning and afternoon hours (except the lunch break). Mean hourly indoor PM_1 levels increased by 15-20% as a result of indoor emissions. On average, the chemical composition of indoor PM_1 was dominated by mineral matter due to re-suspension processes.

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Figure captions

Figure 1. Correlation between $PM_{2.5}$ concentrations obtained by means of high-volume filter samples and laser spectrometers (GRIMM) in the indoor and outdoor environments.

Figure 2. Mean hourly indoor and outdoor BC and PM_1 levels monitored between 17/07/2010 and 26/07/2010.

Figure 3. Seasonal evolution of the sulphate and nitrate contents of indoor and outdoor $PM_{2.5}$ aerosols.

Figure 4. Time series of total indoor BC concentrations (measured), outdoor BC infiltration (calculated) and indoor BC emissions (calculated).

Figure 5. Daily cycle of outdoor, total indoor and indoor emissions for BC (ng/m^3) and PM_1 (µg/m^3).

Figure 6. Indoor BC and PM_1 emissions during work days (Monday to Friday) and on weekends (Saturday-Sunday).

Tables

Table 1. Mean, minimum and maximum levels of black carbon (BC) and PM_1 at the indoor and outdoor locations, for the entire monitoring period. I/O: indoor/outdoor ratio. Ratios calculated on a sample-by-sample basis, not as the ratio of the mean values.

BC	Mean	Min.	Max.
Indoor (ng/m^3)	1572	178	10740
Outdoor (ng/m^3)	1871	115	10060
I/O	0.97	0.14	3.42
PM_1			
Indoor (µg/m^3)	7.6	1.9	55.1
Outdoor (µg/m^3)	12.5	1.3	61.3
I/O	0.76	0.21	6.0

Table 2. BC and PM_1 penetration ratios. S.d.: standard deviation.

	Outdoor/Indoor ratios		
Dates (weekends)	**Mean**	**Min.**	**Max.**
BC			
1-2/05/2010	1.30	0.45	2.61
10-11/07/2010	1.20	0.38	2.30
24-25/07/2010	1.35	0.58	3.23
Mean BC O/I	**1.29±0.08 (s.d.)**		
PM_1			
3-4/04/2010	2.13	0.98	3.57
15-16/05/2010	1.98	0.76	3.04
29-30/05/2010	1.75	1.00	3.23
12-13/06/2010	1.46	0.75	2.87
19-20/06/2010	1.34	0.44	2.85
10-11/07/2010	1.14	0.74	1.45
24-25/07/2010	1.23	0.72	1.65
Mean PM_1 O/I	**1.95±0.38 (s.d.)**		

Table 3. Mean chemical composition (µ/m^3) of indoor PM2.5 over 24-hr periods and during the day (6:00-17:00 UTC) and night (18:00-5:00 UTC) shifts. SIA: secondary inorganic aerosols (sum of sulphate, nitrate and ammonium).

	Daily mean	**Day shift**	**Night shift**
CO_3^{2-}	1.4	1.9	0.9
SiO_2	2.9	3.8	2.5
Al_2O_3	1.0	1.3	0.8
Ca	0.9	1.2	0.6
K	0.5	0.6	0.5
Na	1.0	1.1	0.9
Mg	0.1	0.2	0.1
Fe	0.3	0.3	0.3
P_2O_5	0.1	0.2	0.1
SO_4^{2-}	2.2	2.1	2.7
NO_3^-	0.9	1.1	1.2
Cl	3.2	5.0	2.7
NH_4^+	0.9	1.0	1.0
BC	1.6	2.0	1.4
Mineral	7.2	9.4	5.8
NA+Cl^- (marine)	4.2	6.1	3.7
SIA	4.0	4.2	4.9

Figures1-6

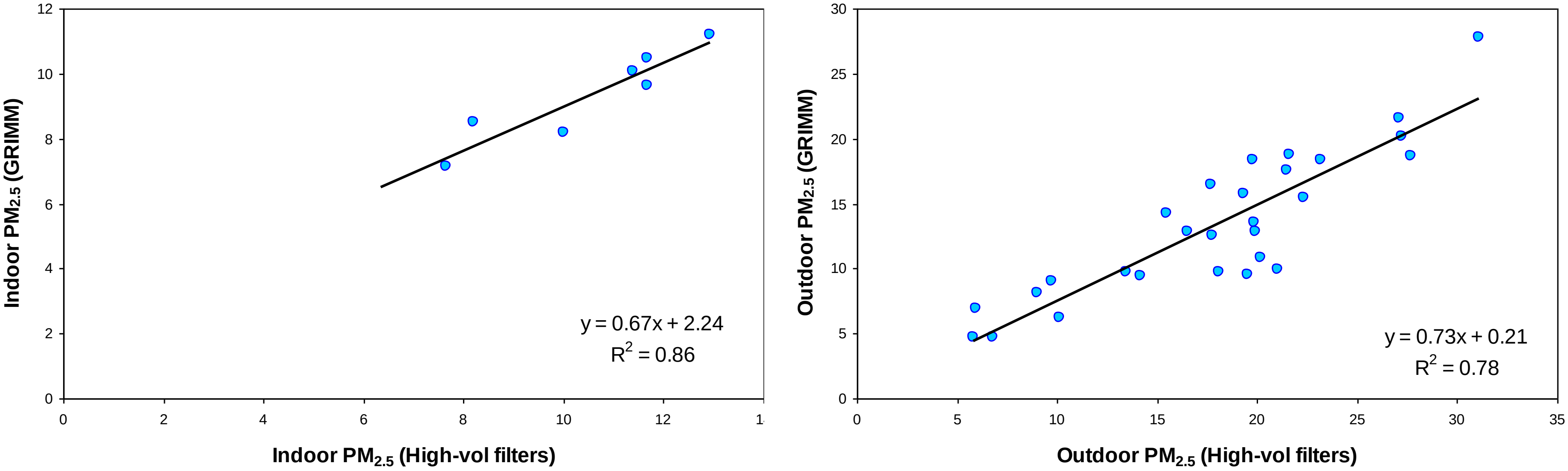

Figure 1

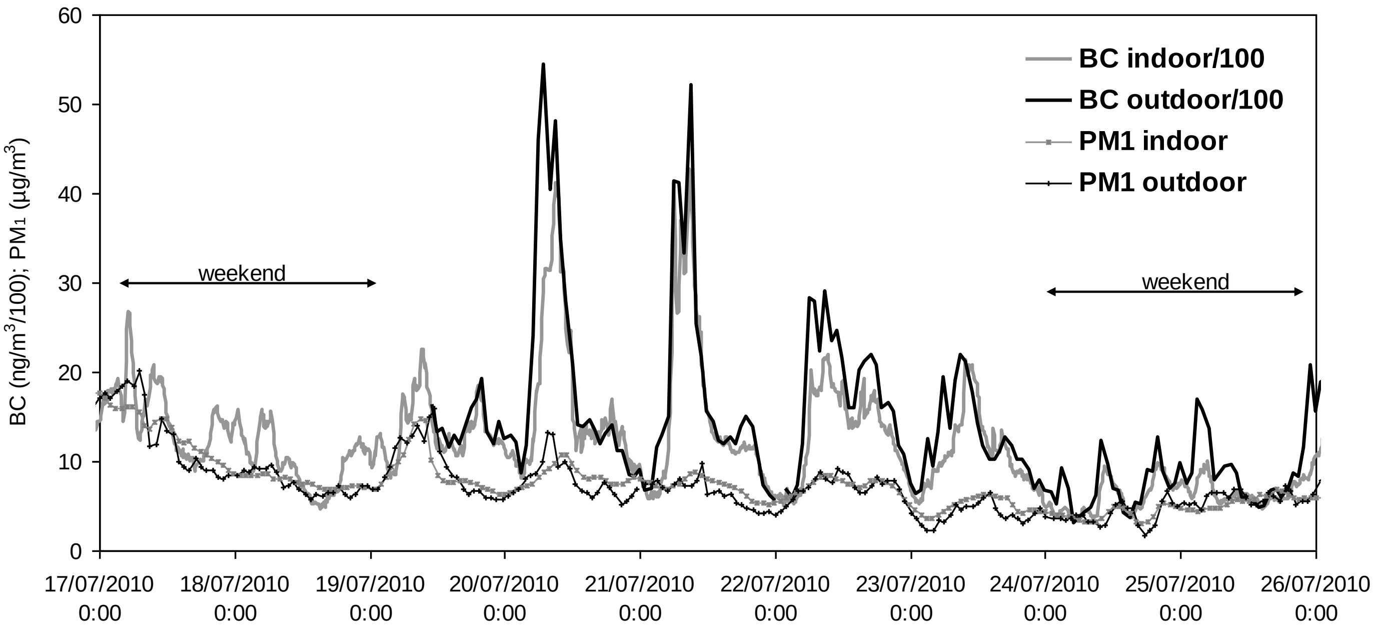
BC indoor/100
BC outdoor/100
PM1 indoor
PM1 outdoor
weekend
weekend
BC (ng/m3/100); PM1 (µg/m3)
60
50
40
30
20
10
0
17/07/2010 0:00
18/07/2010 0:00
19/07/2010 0:00
20/07/2010 0:00
21/07/2010 0:00
22/07/2010 0:00
23/07/2010 0:00
24/07/2010 0:00
25/07/2010 0:00
26/07/2010 0:00

Figure 2

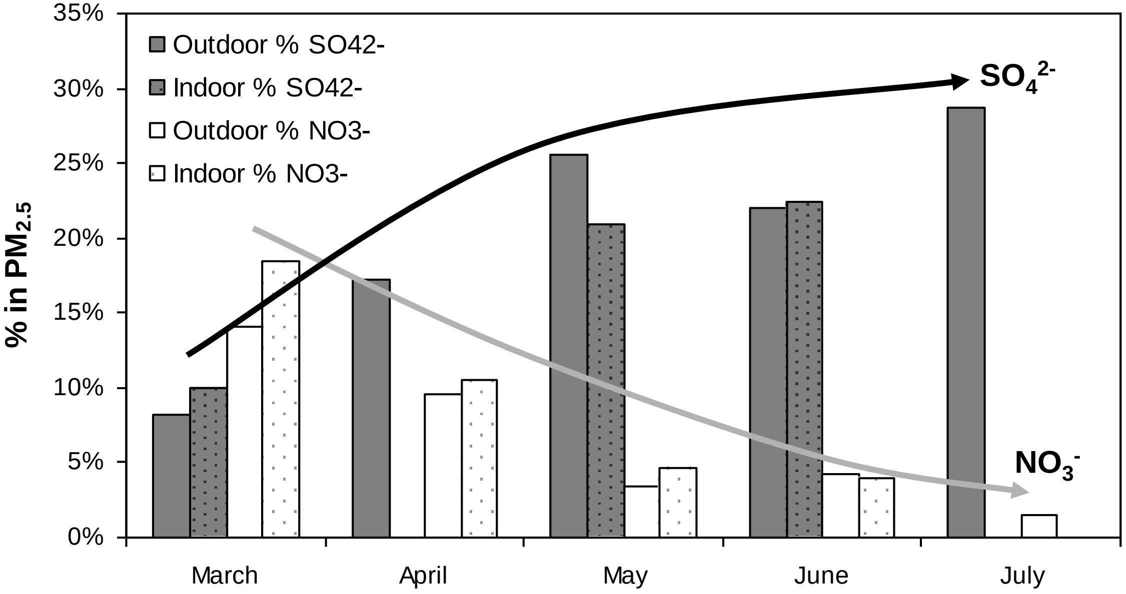
35%
30%
25%
20%
15%
10%
5%
0%
% in PM2.5
Outdoor % SO42-
Indoor % SO42-
Outdoor % NO3-
Indoor % NO3-
SO4 2-
NO3 -
March
April
May
June
July

Figure 3

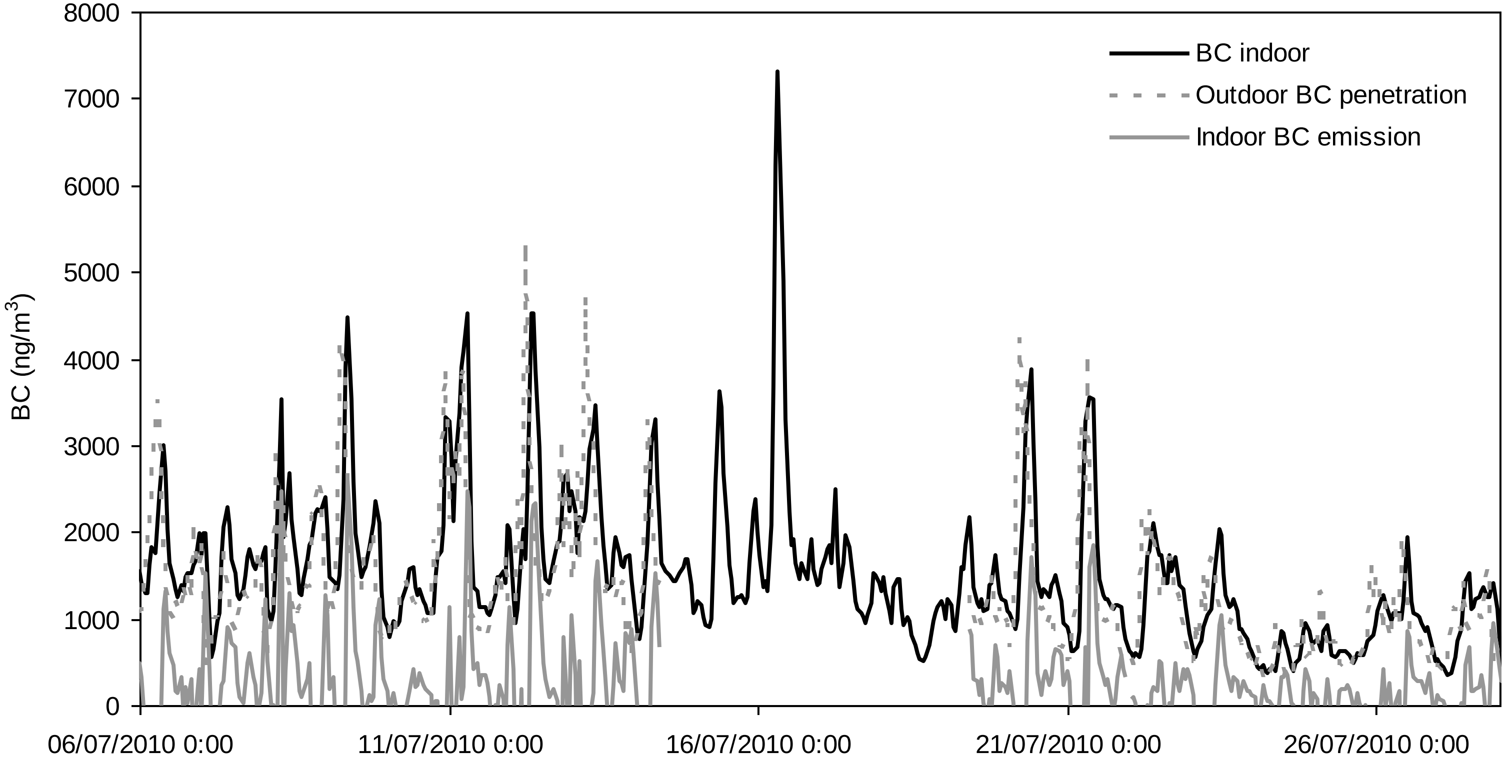
8000
7000
6000
5000
4000
3000
2000
1000
0
BC (ng/m3)
06/07/2010 0:00
11/07/2010 0:00
16/07/2010 0:00
21/07/2010 0:00
26/07/2010 0:00
BC indoor
Outdoor BC penetration
Indoor BC emission

Figure 4

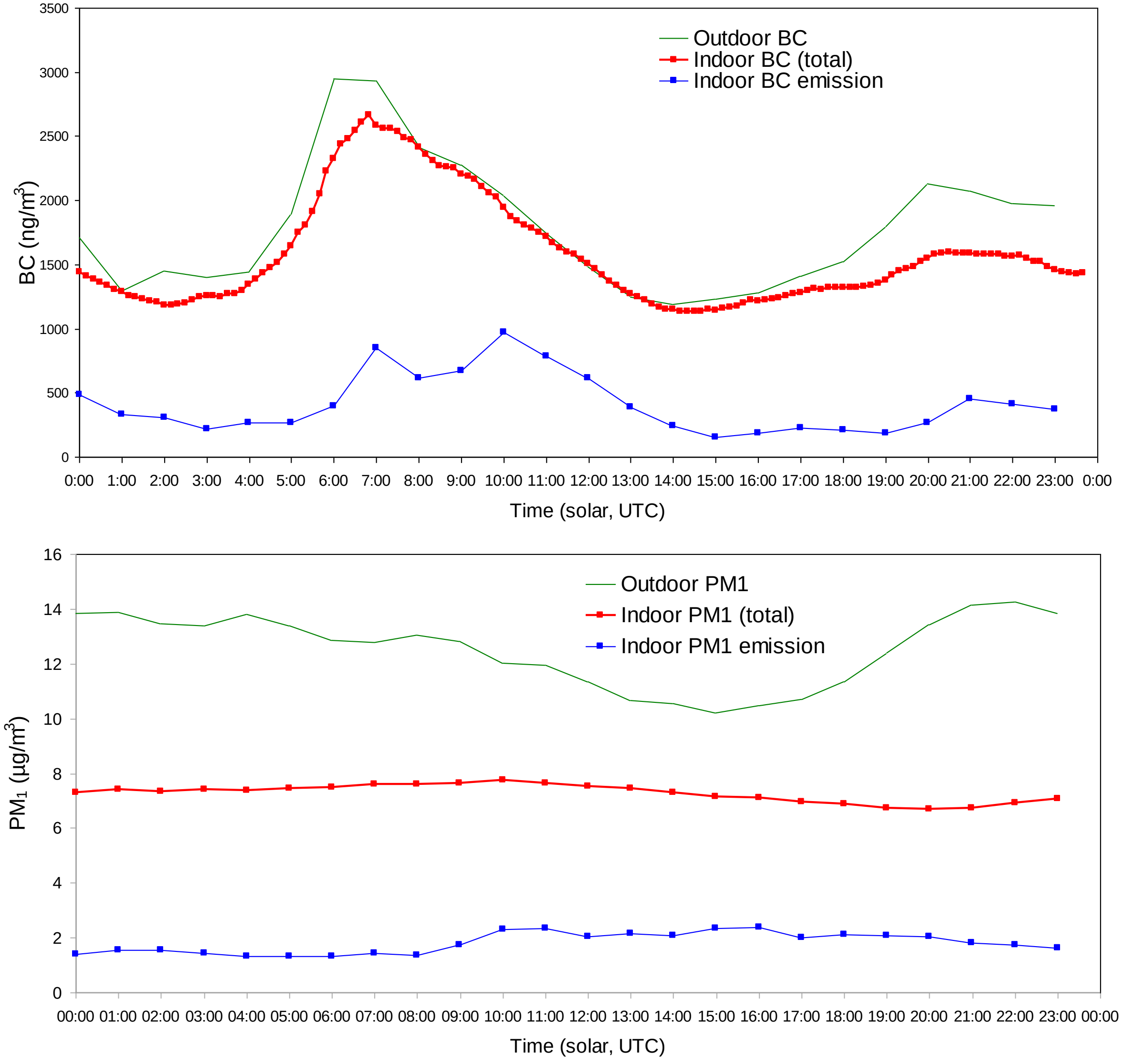
3500
3000
2500
2000
1500
1000
500
0
BC (ng/m^3)
Outdoor BC
Indoor BC (total)
Indoor BC emission
0:00 1:00 2:00 3:00 4:00 5:00 6:00 7:00 8:00 9:00 10:00 11:00 12:00 13:00 14:00 15:00 16:00 17:00 18:00 19:00 20:00 21:00 22:00 23:00 0:00
Time (solar, UTC)
16
14
12
10
8
6
4
2
0
PM$_1$ (µg/m^3)
Outdoor PM1
Indoor PM1 (total)
Indoor PM1 emission
00:00 01:00 02:00 03:00 04:00 05:00 06:00 07:00 08:00 09:00 10:00 11:00 12:00 13:00 14:00 15:00 16:00 17:00 18:00 19:00 20:00 21:00 22:00 23:00 00:00
Time (solar, UTC)

Figure 5

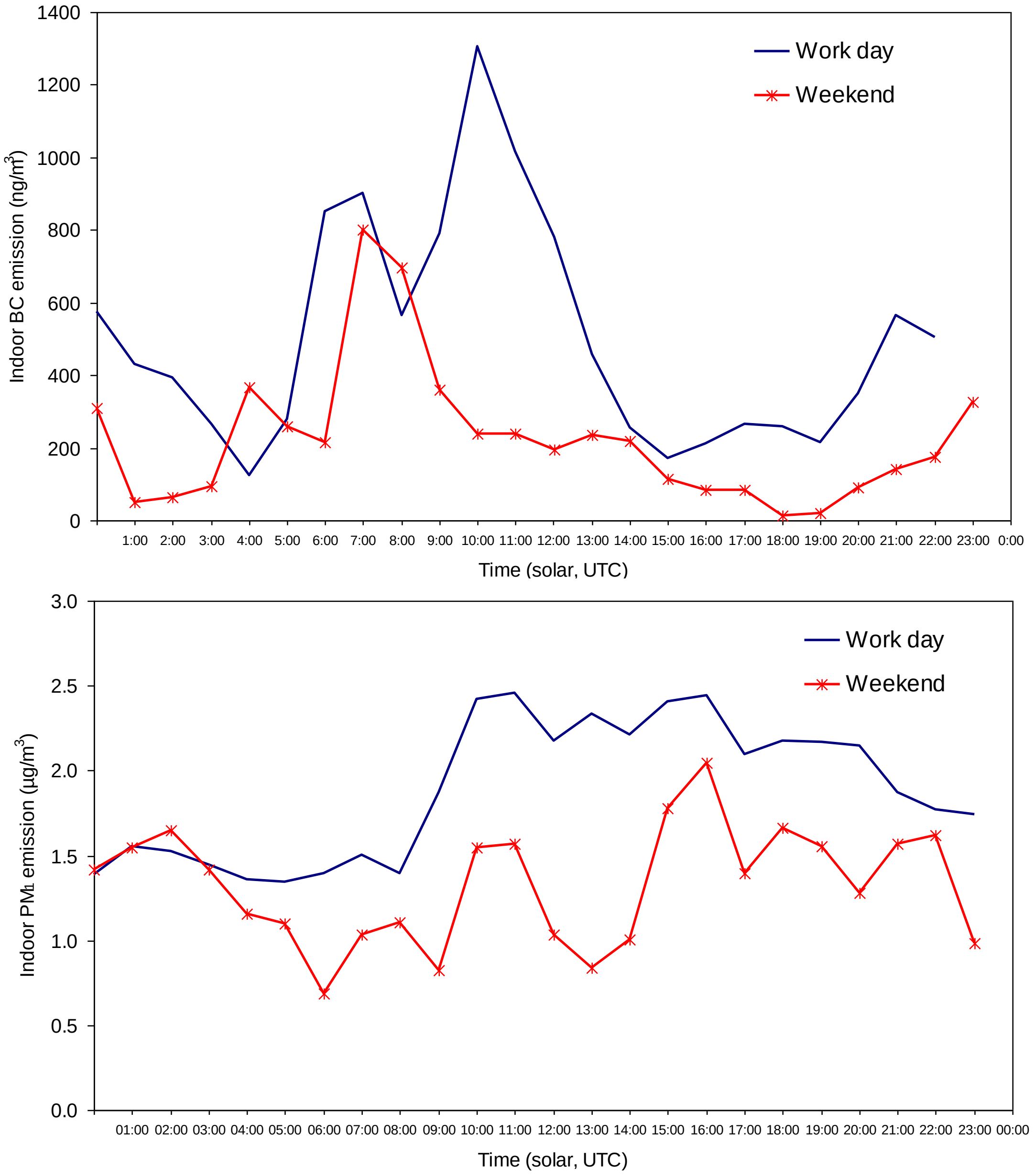
Indoor BC emission (ng/m3)
1400
1200
1000
800
600
400
200
0
1:00 2:00 3:00 4:00 5:00 6:00 7:00 8:00 9:00 10:00 11:00 12:00 13:00 14:00 15:00 16:00 17:00 18:00 19:00 20:00 21:00 22:00 23:00 0:00
Time (solar, UTC)
Work day
Weekend
Indoor PM1 emission (µg/m3)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
01:00 02:00 03:00 04:00 05:00 06:00 07:00 08:00 09:00 10:00 11:00 12:00 13:00 14:00 15:00 16:00 17:00 18:00 19:00 20:00 21:00 22:00 23:00 00:00
Time (solar, UTC)
Work day
Weekend

Figure 6