Workplace exposure to traffic-derived nanoscaled particulates

M Viana, S Díez, A Alastuey, X Querol and C Reche
Institute for Environmental Assessment and Water Research (IDAEA-CSIC), C/ Jordi Girona 18, 08034 Barcelona, Spain.

E-mail: mar.viana@idaea.csic.es

Abstract. Workplace exposure to traffic-derived nanoscaled particulates was determined at a chemical research facility. Sub-micron particles were monitored by means of a multi-angle absorption photometer (MAAP) and a laser spectrometer (GRIMM 1107), providing 10-minute black carbon (BC) concentrations and 15-minute PM$_{1}$ concentrations, respectively, over a 4-month period (22/03/2010 - 28/07/2010). BC levels were simultaneously monitored during 1-day periods using a handheld aethalometer (Magee AE51), with excellent agreement between both techniques (MAAP and AE51, r$^2$=0.96, y=0.84x). The studied laboratory is located on the 5th floor of an 8-storey building in an urban background environment in Barcelona, Spain. The laboratory was not in use during the study period, and both of its doors were kept open at all times in order to ensure air circulation between the study laboratory and the remaining offices and laboratories on the same floor (where workers were exposed). Windows were kept closed at all times. Indoor BC and PM$_{1}$ concentrations were compared with ambient BC and PM$_{1}$ levels from an outdoor monitoring station located at <150 m away from the research facility. Results evidenced the major impact of outdoor vehicular traffic emissions on the levels of nanoscale particulates monitored in the workplace, with clear daily cycles coinciding with traffic rush hours, especially during week days. Penetration ratios were calculated for BC which showed that, even ensuring that all windows were closed, at least 82% of indoor BC concentrations originate from outdoor emissions. Outdoor/indoor penetration ratios were stable for BC (ranging between 1.20 and 1.35) but not for PM$_{1}$ (1.76 to 1.02), suggesting that it is necessary to monitor the variability of penetration factors as a function of time. BC emission sources in the workplace still need to be determined, but could be related to printer/photocopier toner emissions and laboratory work. Potential contamination due to the monitoring instruments (pumps) was discarded through the analysis of daily indoor BC cycles.

1. Introduction
Numerous research works available in the literature have reported significant health risks associated with exposure to particulate matter (PM) [1]. In recent years research interest has been directed towards fine and ultrafine particles (particulates with aerodynamic diameter <1 μm). Epidemiological studies [2] have indicated that fine and ultrafine may have a greater potency to cause adverse health effects than larger particles, given that they have shown increased deposition rates in the lower respiratory tract [3].

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 [4]. Whereas data on outdoor PM levels and chemical composition are widely available in Europe [5], relatively few data are available in the literature regarding the
levels and characteristics of atmospheric particulates in indoor air. This is generally attributed to the
complexity in the determination of PM levels indoors, given the broad variability of direct indoor
sources (cleaning, cooking, smoking, etc.) as well as the multiplicity of microenvironments
(households, work environments, commerce, etc.). Studies [6] have shown that measured particle
levels in indoor environments are greatly influenced by indoor activities, leading to high-concentration
but short-term events. The examination of short-term variations in particulate concentrations is,
therefore, essential in exposure studies.

Assessment of the risk to the community resulting from exposure to airborne pollutants should
ideally include measurements of concentration levels of the pollutants in all microenvironments where
people spend their time. However, due to the vast range of different microenvironments it is not
possible to conduct measurements in all of them. Over the past several decades, exposure to indoor air
pollutants in work environments is believed to have increased due to a variety of factors [7], including
the construction of more tightly sealed buildings, reduced ventilation rates to save energy, the use of
synthetic building materials and furnishings, and the use of chemically formulated cleaning products.
In work environments, indoor air quality problems can be traced to sources including office
equipment, office supplies, construction materials (renovations and new furnishings), maintenance
work (cleaning products, etc.) and outdoor sources.

In this respect, the influence of outdoor PM sources on indoor environments is a key issue to be
taken into consideration. In recent years, indoor/outdoor (I/O) relationships have been studied in the
US and Australia with a special focus on the penetration rates of traffic-related particulates [8].
Conversely, fewer studies are available in Europe and they mostly focus on modelling of ultrafine
particulate concentrations in indoor environments [9]. Studies are especially rare in Spain [10].

While the focus of most of the available studies reported has been on particle mass, there is still
very little information on the I/O relationship as a function of the geochemistry, size or sources of
particulate matter. Theoretically, the indoor particle concentration is a function of a number of factors,
the most important of which are the generation rate of particles indoor, the outdoor particle
centration, air exchange rate, particle penetration efficiency from the outdoor to the indoor
environment, and the particle deposition rate on indoor surfaces [11]. However, in practice, it is
usually very difficult to assess the exposure due to the lack of data and information on the correlation
between indoor and outdoor particles, which are building and environment specific. Understanding the
relationship between indoor and outdoor aerosol particles under different environmental conditions is
of importance for improving exposure estimates and in turn for developing efficient control strategies
to reduce human exposure and thus health risk.

Based on the above considerations, the main objective of the present work is to characterise
exposure to indoor particulate matter of outdoor origin in the work environment. Specifically, our
work focuses on traffic-derived aerosols. Taking one step further from the studies currently available
in the literature, this study characterises workplace exposure to aerosols in terms of nano-scaled
particulate mass (<1 micron, PM$_{1}$) and black carbon concentrations.

2. Methodology
Monitoring of indoor PM$_{1}$ and black carbon (BC) concentrations was carried out at a chemical
research facility belonging to the Spanish National Research Council (Institute for Environmental
Assessment and Water Research, IDAEA-CSIC), in the urban area of Barcelona (NE Spain). The
building is >30 years old. The study location was a vacant laboratory located on the 5th floor of the
building, adjacent to other laboratories and offices which were in use on a daily basis. The study
laboratory 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 circulation with the remaining offices and laboratories on the same floor
(where workers were exposed). Windows were kept closed at all times. 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 same instrumentation was 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. PM$_1$ concentrations were monitored by means of two laser spectrometers (GRIMM 1107) with a 15-minute (indoor) and 30-minute (outdoor) time resolution. In addition, in the indoor environment BC levels were also monitored using a portable aethalometer (Magee AE51), simultaneously with the MAAP instrument. 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 these instruments, one low volume PM$_{2.5}$ sampler was located in the indoor environment, which collected 12-hr samples on 47-mm quartz fibre filter substrates. 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). The chemical composition of the particulates collected on the filters (indoor and outdoor) was characterised by means of ICP-MS, ICO-OES, IC and organic and elemental carbon analysis. The results of the chemical characterisation are not included in the present work.

### 3. Results and Discussion

Mean, minimum and maximum levels of BC and PM$_1$ at the indoor and outdoor locations are summarised in Table 1. PM$_1$ levels, determined by laser spectrometry, were underestimated when compared with results obtained by means of the reference gravimetric method (outdoor PM$_1$=18.6 µg/m$^3$; unpublished data). Automatic data should be corrected by comparison with the reference gravimetric data for air quality monitoring purposes (which are not the focus of the present study).

On average, BC and PM$_1$ levels were higher in the outdoor environment than indoors (19% and 37% higher, respectively), with mean levels for the entire monitoring period of 1572 ngBC/m$^3$ and 7.5 µgPM$_1$/m$^3$ indoors, and 1871 ngBC/m$^3$ and 10.3 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 high-concentration but short-duration events described above. Indoor/outdoor (I/O) ratios were calculated on a sample-by-sample basis and then averaged for the entire period, and thus were 0.97 and 0.92 for BC and PM$_1$, respectively, which are much higher than values found in the literature for urban environments (0.60-0.65 for BC and 0.38-0.46 for PM$_1$, [12]). These ratios evidence a major influence from outdoor particles in the indoor environment, probably as a result of inadequate insulation of the building due to its old age (>30 years).

![Table 1](https://example.com/table1.png)

|               | Mean | Min. | Max.  |
|---------------|------|------|-------|
| BC Indoor     | 1572 | 178  | 10740 |
| BC Outdoor    | 1871 | 115  | 10060 |
| BC Indoor/Outdoor | 0.97 | 0.14 | 3.42  |
| PM$_1$ Indoor | 7.5  | 1.9  | 41.5  |
| PM$_1$ Outdoor| 10.3 | 0.7  | 65.0  |
| PM$_1$ Indoor/Outdoor | 0.92 | 0.21 | 6.0   |

The time series of simultaneous indoor and outdoor PM$_1$ and BC concentrations were analysed in search for emission patterns. As shown in Figure 1, the correlation between indoor and outdoor measurements was significantly high, with good correspondence in absolute values ($y=1.07x$) but 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 (06:00-08:00 and 18:00-20:00 UTC), 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 major and common emission source in both environments.
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BC (ng/m$^3$/100); PM$_1$ (µg/m$^3$)

Figure 1. Indoor BC and PM$_1$, and outdoor BC levels, monitored in a chemical research facility.

In order to quantify the influence of outdoor vehicular emissions of PM$_1$ and BC, penetration factors were calculated based on the methodology described in [13]. This methodology aims to determine the penetration ratio of outdoor particles into the indoor environment, in order to be able to determine the indoor particle load which is originated by indoor emissions, as opposed to the particles generated outdoors but infiltrated and detected indoors. To this end, outdoor-to-indoor ratios (O/I) are calculated for time periods during which no indoor PM emissions are generated, typically weekends or Sundays in industrial facilities and workplaces. In the chemical research facility under study, weekends were representative of periods with zero indoor-generated emissions. Penetration ratios for BC are shown in Table 2, calculated for the 3 weekends for which simultaneous indoor and outdoor BC data were available. According to our results, the O/I ratio remained relatively constant between May and July, with a mean value of 1.29 (ranging between 1.20 and 1.35, standard deviation = 0.08), suggesting that the infiltration of BC particles was relatively constant and independent of the season (spring vs. summer). Conversely, the O/I ratios calculated for PM$_1$ showed a decrease from April to July (Table 2, 7 weekends available for calculation of the ratios for PM$_1$). Whereas the mean O/I ratio for PM$_1$ was 1.76 in April, it was close to 1 (0.95-1.02) in July, indicating a much higher degree of particle infiltration during the summer period than in spring. Taking into account that windows remained closed at all times, this increase in particle infiltration can only be interpreted as a change in particle characteristics, either particle composition and/or size. Particulate sulphate formation is enhanced in summer due to faster oxidation rates of SO$_2$, which results in higher concentrations of fine sulphate particles in summer [14]. One possible interpretation for the different O/I ratios obtained for BC and PM$_1$ could be the increase in sulphate particles in ambient air, which would result in a finer grain size distribution of the PM$_1$ fraction and thus an enhanced infiltration from outdoors to the indoor environment. In addition, this would not influence the penetration ratio of BC particles, with a grain size distribution which is unaffected by seasonal trends. The chemical speciation data of PM$_{2.5}$ particles collected on filters (not included in the present work) will provide further insights into this issue.

Consequently, our results evidence that penetration factors do not depend only on the physical barriers (e.g., windows) 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 of indoor particles originating from indoor and outdoor emissions, it is necessary to monitor the variability of the calculated penetration factors as a function of time.
Table 2. BC and PM$_1$ penetration ratios.

|                  | Outdoor/Indoor 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)**   |      |      |
| **PM$_1$**       |                    |      |      |
| 3-4/04/2010      | 1.76               | 0.81 | 2.95 |
| 15-16/05/2010    | 1.63               | 0.66 | 2.44 |
| 29-30/05/2010    | 1.44               | 0.83 | 2.68 |
| 12-13/06/2010    | 1.20               | 0.62 | 2.37 |
| 19-20/06/2010    | 1.20               | 0.32 | 2.66 |
| 10-11/07/2010    | 0.95               | 0.60 | 1.29 |
| 24-25/07/2010    | 1.02               | 0.57 | 1.39 |
| **Mean PM$_1$ O/I** | **1.61 (±0.30)** |      |      |

Indoor BC emissions were then estimated based on the penetration factors described above and the outdoor and indoor BC concentrations [13]. For the sake of brevity, results are only shown for BC. Figure 2 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, 82% of the BC concentrations measured in the indoor environment were originated outdoors, and only 18% were linked to indoor BC emissions. Even after the subtraction of indoor BC concentrations of outdoor origin, indoor BC emissions still showed peaks coinciding with vehicular traffic rush hours (Figure 2). This is probably a consequence of the application of mean weekend O/I ratios to hourly data. The daily cycle of total indoor BC concentrations (Figure 3) showed a clear traffic-derived trend for total indoor BC, whereas indoor BC emissions of indoor origin showed one major increase between 9:00-12:00 UTC, probably coinciding with characteristic workplace activities linked to indoor BC sources (printing, photocopying, etc). The potential contamination due to the monitoring instruments (pumps) was discarded based on the clear daily cycles observed. In addition, the influence of outdoor traffic-derived aerosols was also detected as a secondary BC peak at 7:00 UTC. Once again, this indicates that particles infiltrated from the outdoor environment cannot be fully segregated by applying O/I ratios. When the daily cycle of indoor BC emissions was analysed separately for working days and weekends (Figure 4), the morning traffic peak was detected throughout the entire week (with different intensities) given that traffic emissions only decrease on weekends. Conversely, the 9:00-12:00 UTC peak (due to workplace activities) was only present during weekdays, and it was completely absent on weekends when the building was empty. In terms of mass, the BC increase due to workplace activities represents a 71% increase (616 ng/m$^3$) on weekdays with respect to weekends (on average 254 ng/m$^3$ on weekends and 870 ng/m$^3$ on weekdays, for the period 09:00-13:00 h).
Figure 2. Indoor BC concentrations measured, and indoor BC emissions inferred from calculated BC penetration ratios.

Figure 3. Daily cycle of total indoor BC concentrations and indoor BC concentrations of indoor origin.
Finally, semi-hourly BC data obtained with the MAAP instrument were compared with the data obtained with the mini aethalometer AE51 (Figure 5), for the purpose of validating the results provided by the handheld instrument. Results showed that 24 hours is the maximum monitoring period in order to obtain an acceptable comparison between the handheld and the online instruments, even if the battery life of the AE51 instrument is longer. Monitoring times >24h with the handheld instrument resulted in filter saturation which decreased the correlation dramatically ($r^2=0.96$ for <24h periods vs. $r^2=0.61$ for >24h).

Figure 5. Comparison between 10-minute BC data obtained by means of an online absorption photometer (MAAP) and a handheld aethalometer (AE51).

4. Conclusions
Workplace exposure to nanoscaled particulates of indoor and outdoor origin was determined at a chemical research facility by means of a multi-angle absorption photometer (MAAP) and a laser spectrometer (GRIMM 1107), providing black carbon (BC) and PM1 concentrations. Indoor BC and PM1 concentrations were compared with ambient BC and PM1 levels from an outdoor monitoring station located at <150m away from the research facility. Results evidenced the major impact of
outdoor vehicular traffic emissions on the levels of nanoscale particulates monitored in the workplace, with clear daily cycles coinciding with traffic rush hours, especially during week days. Penetration ratios of 1.29 (with 0.08 standard deviation) and 1.61 (with 0.30 standard deviation) were obtained for BC and PM$_{1}$, respectively. While no seasonal trend was observed for these ratios for BC, they decreased strongly for PM$_{1}$ from March to July (1.76 to 1.02). This indicates a higher degree of particle infiltration during the summer period than in spring for PM$_{1}$ (not for BC), which can only be interpreted as a change in particle characteristics, either particle composition and/or size. One possible interpretation for the different O/I ratios obtained for BC and PM$_{1}$ could be the increase in sulphate particles in ambient air, which would result in a finer grain size distribution of the PM$_{1}$ fraction and thus an enhanced infiltration from outdoors to the indoor environment. Even ensuring that windows were closed at all times, at least 82% of indoor BC concentrations originated from outdoor emissions due to inadequate insulation of the building materials. BC emission sources in the workplace still need to be determined, but should be related to printer/photocopier toner emissions and laboratory work.

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