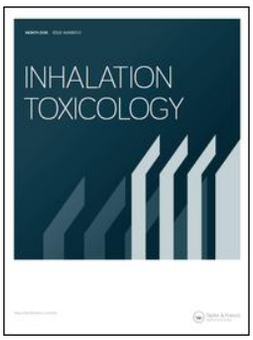

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MEASUREMENT TECHNOLOGY

Monitoring exposure to airborne ultrafine particles in Lisbon, Portugal

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Abstract

The aim of this study is to contribute to the assessment of exposure levels of ultrafine particles (UFP) in the urban environment of Lisbon, Portugal, due to automobile traffic, by monitoring lung-deposited alveolar surface area (resulting from exposure to UFP) in a major avenue leading to the town centre during late Spring, as well as in indoor buildings facing it. This study revealed differentiated patterns for week days and weekends, consistent with $PM_{2.5}$ and PM_{10} patterns currently monitored by air quality stations in Lisbon. The observed ultrafine particulate levels could be directly related with the fluxes of automobile traffic. During a typical week, UFP alveolar deposited surface area varied between 35.0 and 89.2 $\mu m^2/cm^3$, which is comparable with levels reported for other towns such in Germany and United States. The measured values allowed the determination of the number of UFP per cm^3, which are comparable to levels reported for Madrid and Brisbane. In what concerns outdoor/indoor levels, we observed higher levels (32–63%) outdoor, which is somewhat lower than levels observed in houses in Ontario.

Keywords: Airborne ultrafine particles, urban environment, indoor/outdoor

Introduction

The adverse health effects of particulate matter have been reported in numerous scientific studies (Pope & Dockery, 2006; Buonanno et al., 2010). A number of epidemiological studies also associated these effects with particle mass concentration including $PM_{2.5}$ (Pope, 2000) and PM_{10} (Loomis, 2000), as well as ultrafine particle (UFP), number concentration (Hauser et al., 2001), surface area concentration (Driscoll, 1996), and overall exposure rate (Siegmann & Siegmann, 1998). Apart from that, a number of studies worldwide have found an elevated risk of lung and ovarian cancer associated with exposure to diesel engine fumes and other emissions derived from automobile traffic (Ramachandran et al., 2005). An important information gap, that limits the use of this data for epidemiological studies and risk assessment evaluations, is the absence of quantitative exposure data from which to estimate the dose-response relationship (Mauderley, 1992). It is not clear, at the moment, which fraction of the particulate matter is responsible for the health effects. Nearly all of the mass emitted by engines is in the fine particle range and nearly all the number is in the nanoparticle range (Kittelson, 1992). Several studies have suggested that, at similar mass concentrations, nanometer size particles are more harmful than micron size particles (Seaton et al., 1995; Oberdörster, 1996). One possible cause for this might be that, since the number of particles and also particle surface area per unit mass increases with decreasing particle size, and also as pulmonary deposition increases with decreasing particle size, the dose by particle number or the surface area will increase as the particle

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size decreases (Ramachandran et al., 2005). Current workplace and ambient air environmental exposure limits, that have been established long ago, are based on particle mass. However, this criterion does not seem adequate in what concerns UFP as these materials are, in fact, characterized by very large surface areas, which has been pointed out as the distinctive characteristic that could even turn out an inert substance into a toxic substance, having the same chemical composition, but exhibiting very different interactions with biological fluids and cells (Pope & Dockery, 2006). Therefore, it seems that, assessing human exposure based on the mass concentration of particles, which is widely adopted for particles over 1 μm, would not be adequate in this particular case. In fact, UFP have far more surface area for the equivalent mass of larger particles, which increases the chance that they may react with body tissues (Buonanno et al., 2010). Thus, a growing number of experts (Pope, 2000; Loomis, 2000) have been claiming that surface area should be used for UFP exposure and dosing. As a result, assessing workplace conditions and personal exposure based on the measurement of particle surface area is of increasing interest. It is well-known that lung deposition is one of the most efficient ways for airborne particles to enter the body and potentially cause adverse health effects. If UFP can deposit in the lung and remain there, have an active surface chemistry and interact with the body, then, there is a strong potential for exposure and dosing. (Oberdörster, 1996) showed that surface area plays an important role in the toxicity of nanoparticles, and this is the measurement metric that best correlates with particle-induced adverse health effects. The potential for adverse health effects is directly proportional to particle surface area (Driscoll, 1996). Although the mass concentration of fine particles, namely in the ranges of $PM_{2.5}$ and PM_{10}, has been currently monitored in urban environments in large towns worldwide, very few studies have been made regarding UFP, as referred in Table 1. The majority of these studies report concentrations in terms of number of particles per cm^3, while some other groups measured UFP concentration in terms of surface area, expressed as $\mu m^2/cm^3$, which we feel is the most significant metric in what concerns this type of pollutant, particularly bearing in mind the existing relationship between surface area of UFP and its health effects. Regarding indoor concentrations some studies were focused on the determination of fine particles concentration, expressed as $\#/cm^3$, as described also in Table 1, but none of these studies comprised the determination of surface area of indoor particles.

The primary aim of this study is to contribute to the assessment of exposure levels of UFP in the urban environment of Lisbon, Portugal, due to automobile traffic, by monitoring lung-deposited alveolar surface area (resulting from exposure to UFP) in a major avenue leading to the town centre during late Spring, as well as in indoor buildings facing it.

Table 1. Previous studies measuring outdoor and indoor levels of fine and ultrafine particles.

Sampling sites	Particle sizes sampled	Measuring units	Outdoor/ indoor	References
Beijing, Paris, Mexico City, New York, Tokyo, Zurich	>1 μm	$\#/cm^3$	Outdoor	Zhiqiang et al., 2000
Birmingham, UK	<10 μm	$\#/cm^3$	Outdoor	Shi et al., 2001
Brisbane, Australia	Ultrafine particles (UFP)	$\#/cm^3$	Outdoor	Morawska et al., 2002
Madrid, Spain	UFP	$\#/cm^3$	Outdoor	Gomez-Moreno et al., 2011
Minneapolis, USA	UFP	$\mu m^2/cm^3$	Outdoor	Ramachandran et al., 2005
Dusseldorf, Germany	UFP	$\mu m^2/cm^3$	Outdoor	Kuhlbusch et al., 2004
Los Angeles, USA	UFP	$\mu m^2/cm^3$	Outdoor	Ntziachristos et al., 2007
Espoo, Finland	UFP	$\#/cm^3$	Outdoor and indoor	Hussein et al., 2006
Helsinki, Athens, Amsterdam, Birmingham	UFP	$\#/cm^3$	Indoor	Hoek et al., 2008
Windsor, Canada	UFP	$\#/cm^3$	Outdoor and indoor	Kearney et al., 2011

Methods

Sampling site

Measurements were made in 56 consecutive days on late spring, from the 4th of April until the 30th of May, 2011, on a trailer which was located downwind on a 4-lane avenue in Lisbon, Portugal, shown in Figure 1, which is one of the main accesses to the town center coming from West (Benfica) and its surroundings. The measurements were made during the dry season, in the absence of rain. Lisbon has a subtropical-mediterranean climate with mild winter and warm to hot summers. The annual average temperature is 17°C, 21°C during the day and 13°C at night, with an annual precipitation of 725.8 mm mainly concentrated in autumn and winter months. Summer lasts about 6 months, from May to October. In the measuring period, the average daily temperature was 17.4°C, with a maximum temperature of 21.4°C during day and a minimum temperature of 13.3°C at night. The consistency of this weather on a daily basis ensures that representative results were obtained during the duration of this study. Previous studies focused on air pollution episodes in Lisbon area have characterized their driving meteorological conditions as well as their typical atmospheric patterns: in summer the situation permits the development of mesoscale flows land-sea breeze and upslope-downslope (anabatic-katabatic) flow (Barros et al., 2003), leading to the occurrence of episodic air pollution events. Thus, the worst pollution

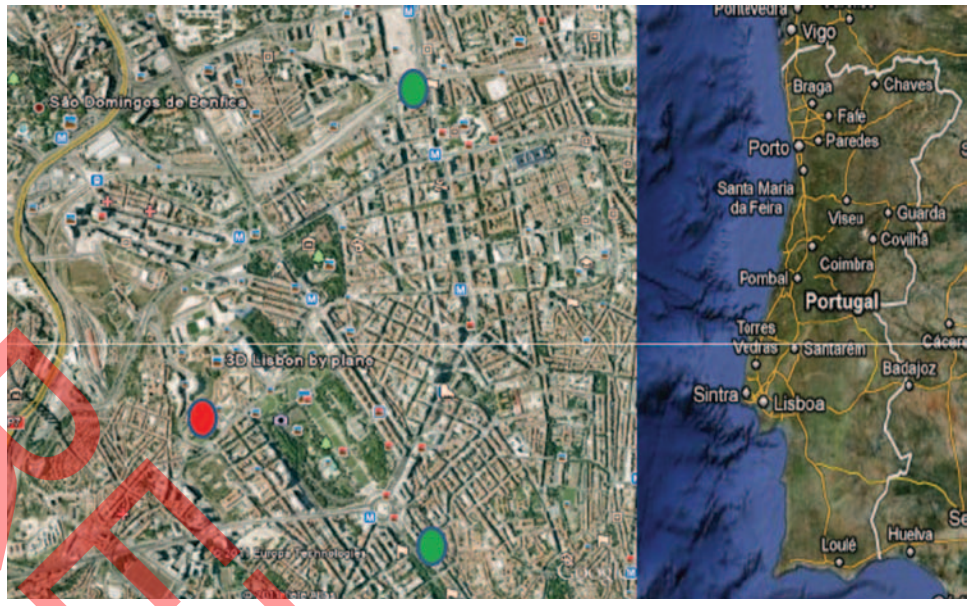

Figure 1. Location of sampling site in Lisbon (red dot indicates sampling site and green dots indicates Entrecampos and Av. Liberdade air quality stations).

episodic situations in this region usually coincide with typical summer weather. During the sampling periods no air pollution episodes took place, and no significant Eastern Atlantic Ocean influence occurred in the sampling site as it is located in the middle of the town, surrounded by tall buildings. It is estimated that exhaust gases from automobiles account for 44.0% and 11.7% of NOx and volatile organic compounds emissions respectively, in Lisbon area, and 12.5% of PM_{10} (Gois et al., 2009). The contribution of exhaust gas emissions to the concentration levels of UFP has not been quantified previously, but it is expected to be substantial. The automobile traffic fluxes entering and leaving the center of Lisbon have been previously monitored for these main access avenues (Rocha, 2008). It was noticed that the incoming traffic during the morning increases from 400 until 1,000 vehicles/h from 8:00 AM to 9:00 AM. In the afternoon, the opposite flux leaving the town center increases from 800 to 1,000 vehicles/h between 1700 h and 1800 h and sometimes reaches 1,100 vehicles/h at 2000 h, during working days. It is also estimated that this traffic consisted of about 40% of diesel vs. 60% of gasoline-fueled vehicles.

Through this avenue, where measurements have been done, heavy fluxes of traffic enter the town, consisting of automobiles, trucks, and buses. For comparison purposes, the air quality data on particulate emissions ($PM_{2.5}$ and PM_{10}) measured regularly on two nearby station in Entrecampos and Av. da Liberdade were also obtained as part of the information made public by the Portuguese Ministry of Environment. Measurements were made during week days and also at weekends, during 24-h periods, every 10 s. This station is also located in a 4-lane avenue of Lisbon, which has similar traffic patterns of the sampling site. Also, indoor measurements were done inside the room (facing the avenue) of an apartment on the second floor of an eight-stories high building located in the same avenue where other measurements took place. This building is directly facing the site where the trailer was installed, so that we can assume that the sampling location is only one, but situated at two different heights: at ground level (trailer) and at two stories high (indoor). Regarding outdoor, baseline measurements were made inside the trailer.

Measuring equipment

For measuring UFP exposure a Nanoparticle Surface Area Monitor (NSAM), TSI, Model 3550 (Shoreview, MN), was used. This equipment indicates the human lung-deposited surface area of particles expressed as square micrometers per cubic centimeter of air ($\mu m^2/cm^3$), corresponding to tracheobronchial (TB) or alveolar (A) regions of the human lung, according to the ICRP deposition model developed by ACGIH (Phalen, 1999). This equipment is based on diffusion charging of sampled particles, followed by detection of the charged aerosol using an electrometer. Using an integral pump, an aerosol sample is drawn into the instrument through a cyclone with a 1 μm cut point. The sample flow is then split, one stream going through a set of carbon and HEPA filters and an ionizer to introduce positively charged ions into a mixing chamber. The other aerosol flow stream is mixed with the ionized stream in a mixing chamber and charged aerosol and excess ions move onto an ion trap. The ion trap voltage can be set to TB or A response. The ion trap acts as an inlet conditioner or a size-selective sampler for the electrometer, by collecting the excess ions and particles that are not of a charge state, corresponding to the TB or A response settings. The aerosol, then moves on to the electrometer for charge measurement, where current is passed from the particles to a conductive filter and measured by a very sensitive amplifier. The charge

measured by the electrometer is directly proportional to the surface area of the particles passing through the electrometer.

Particle number concentration and size distribution were measured using a Scanning Mobility Particle Size Spectrometer (SMPS), TSI, Model 3034 (Shoreview, MN). The system consists of three components, (i) a bipolar radioactive charger for charging the particles, (ii) a differential mobility analyzer for classifying particles by electrical mobility, and (iii) a condensation particle counter for detecting particles. The SMPS measures Dp (in terms of electrical mobility diameter) between 10 and 487 nm using 54-size channels (32 channels per decade) for number concentrations in the range from 102 to 107 #/cm^3.

Morphological analysis were conducted using a Nanometer Aerosol Sampler, TSI, Model 3089 (Shoreview, MN) to collect particles on a mesh Transmission Electron Microscopy (TEM) copper grid with a carbon/formvar support film, and these samples were analyzed throughaTEM microscope, FEI, model Tecnel G2, 200 kV, Twin Lens.

Results and discussion

Measurement results, over a typical week, are presented in Table 2, which also shows calculated values of time-weighted average (TWA) for 8-h periods, total deposited alveolar area and lung area covered by particle matter. It should be noted that TWA is the average exposure over a specified period of time, usually a nominal 8 h, which is a parameter frequently used in exposure assessments in indoor environments. Although it is usually not frequently used for outdoor environments, it is useful for further comparisons with indoor exposures.

Apart from TWA, Table 2, also shows total deposited alveolar area, which is calculated as the sum of all measured instantaneous deposited alveolar area values during sampling period; and lung area covered by particles which is calculated by dividing the total deposited alveolar area per an average lung area of 80 m^2.

Table 2. Measurement results over a typical week (late Spring, 2011).

Sampling conditions	Average deposited area (μm^2/cm^3)	Range of values (μm^2/cm^3)	TWA for 8 h (μm^2/cm^3)	Total deposited area (μm^2)	Lung area covered by particles (μm^2/m^2)
Baseline	34.1 ± 5.0	25.5–50.3	1.07	5.12×10^5	6.40×10^3
Monday	57.6 ± 5.7	14.7–343.3	172.8	8.29×10^7	1.04×10^6
Tuesday	89.2 ± 8.0	27.4–510.5	267.6	1.28×10^8	1.61×10^6
Wednesday	87.1 ± 7.5	31.1–421.1	261.4	1.25×10^8	1.57×10^6
Thursday	82.2 ± 7.2	23.2–245.7	165.2	4.72×10^7	5.90×10^5
Friday	75.0 ± 6.9	17.8–511.1	143.6	4.70×10^7	4.60×10^5
Saturday	35.0 ± 4.5	6.91–365.5	107.4	4.26×10^7	3.40×10^5
Sunday	34.9 ± 4.0	4.94–252.2	104.7	4.10×10^7	3.20×10^5

TWA, time-weighted average.

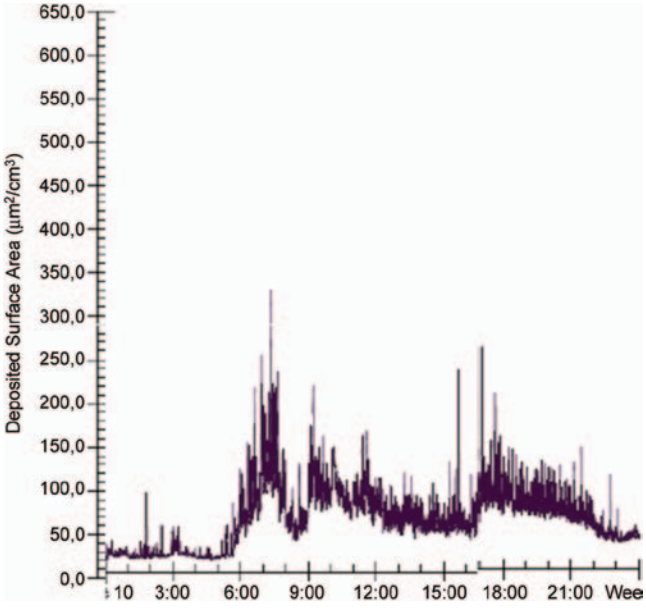

Figure 2. Measurements over a typical week day: Tuesday, the 10th May 2011.

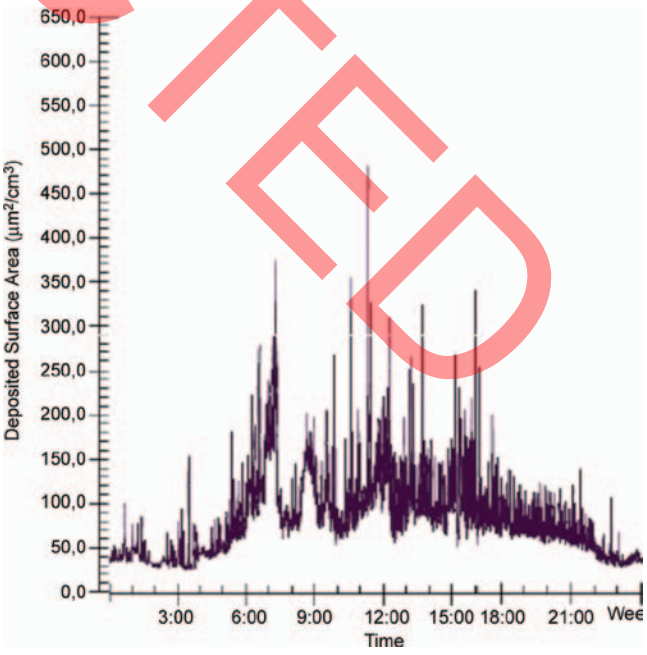

Figure 3. Measurements over a typical week day: Wednesday, the 11th May 2011.

Figure 2 shows the measured alveolar deposited surface area from 0000 h of Tuesday, the 10th May 2011 until 2400 h of the same day, a typical week day. Figure 3 shows the same type of measurements for the following day, Wednesday, the 11th May 2011, from 0000 to 2400 h, another typical week day. Figure 4 shows measurements done from 0000 to 2400 h of Sunday, the 8th May 2011, a typical weekend day.

In selected periods, measurements were made both indoor and outdoor, in terms of A and also TB deposition modes, which are presented in Table 3.

These measurements exhibited differentiated patterns for week days and also for weekend days, as expected, and previously noticed for other urban environments (Morawska et al., 2002; Ntziachristos et al., 2007). As the obtained measurements have shown always similar patterns regarding weekdays and weekend days, other measurements are not shown here.

Figure 5, which shows three superimposed week day patterns (sample 4 = Tuesday, sample 5 = Wednesday, sample 6 = Thursday), shows the similarity of those patterns. On the opposite, Figure 6, which shows superimposed week (sample 3 = Monday, sample 4 = Tuesday, sample

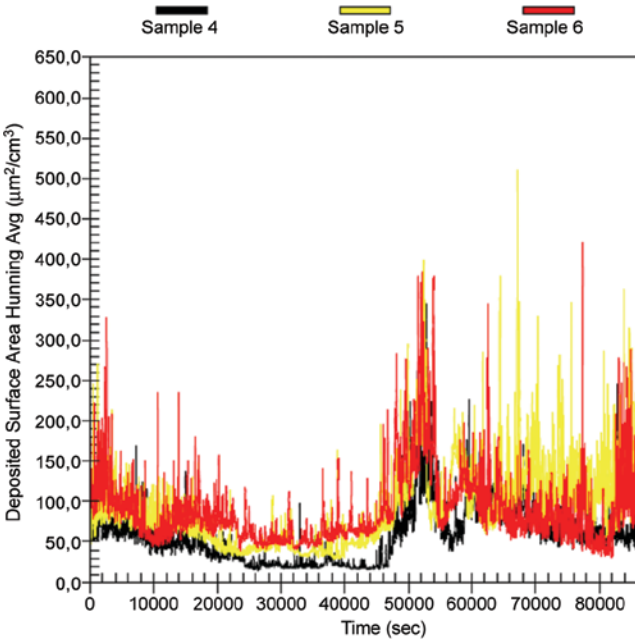

Figure 5. Superimposed measurements for 3 week days.

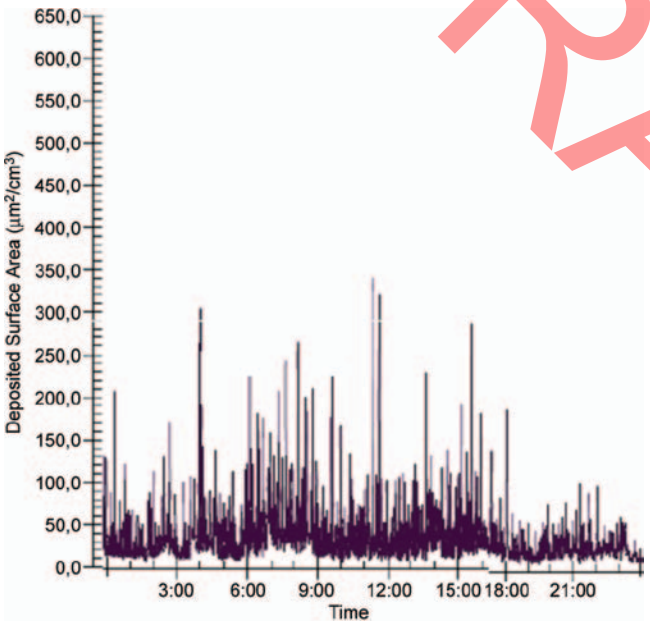

Figure 4. Measurements over a typical weekend day: Sunday, the 8th May 2011.

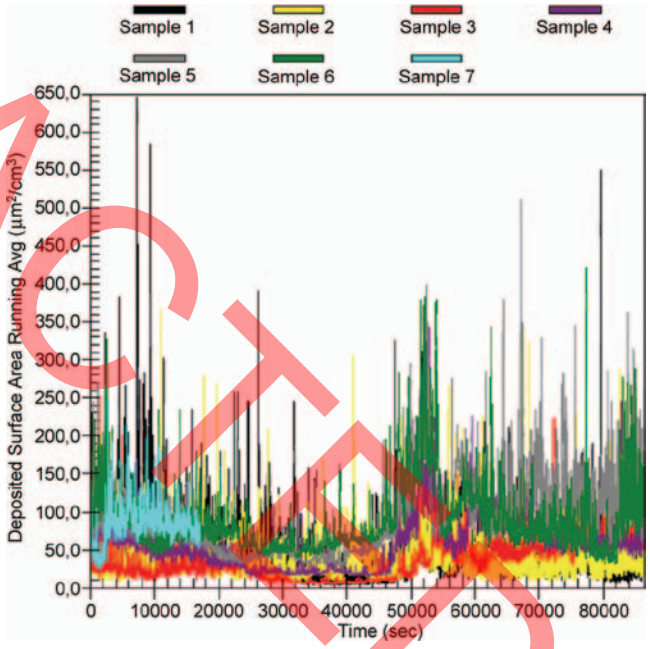

Figure 6. Superimposed measurements for 7 days, both week days and weekend days.

Table 3. Indoor/outdoor measurement results over typical days (late Spring, 2011).

Sampling conditions	Average deposited area ($\mu m^2/cm^3$)	Range of values ($\mu m^2/cm^3$)	TWA for 8 h ($\mu m^2/cm^3$)	Total deposited area (μm^2)	Lung area covered by particles ($\mu m^2/m^2$)
Weekend indoor (TB)	5.7 ± 0.3	5.18–6.29	0.49	2.33×10^5	2.92×10^3
Weekend oudoor (TB)	16.6 ± 9.1	6.91–73.1	1.61	7.74×10^5	9.68×10^3
Weekend indoor (A)	33.1 ± 5.0	24.5–51.3	1.07	5.11×10^5	6.40×10^3
Weekend outdoor (A)	59.2 ± 24.4	17.5–188.7	10.2	4.88×10^6	6.10×10^4
Weekday indoor (A)	29.1 ± 1.0	26.9–31.4	2.49	1.19×10^6	1.49×10^4
Weekday outdoor (A)	108.3 ± 38.1	35.5–239.2	21.4	1.03×10^7	1.29×10^5

A, alveolar; TB, tracheobronchial; TWA, time-weighted average.

5 = Wednesday, sample 6 = Thursday, sample 7 = Friday) and also weekend days (sample 1 = Saturday and sample 2 = Sunday), exhibits considerable differences which can be, of course, attributed to heavier fluxes of automobile traffic during week days when compared to weekend days.

On week days, the diurnal profile of the particle surface concentrations shows that in the early night hours, from 000 to around 0300 h, the concentrations have values in the approximate range of the baseline. Some small peaks can be observed around 0300 h, during a short time (15–30 min) which can be attributed to the traffic fluxes of garbage collection trucks as well as street cleaning vehicles, both operated on diesel fuel, operating at this period. As particle surface concentrations are clearly influenced by automobile traffic, concentrations clearly start to increase from 0500 to 0600 h, thus reflecting traffic entering the center of the town. At these early hours, traffic is mostly constituted by trucks and buses largely fueled by diesel. This traffic fluxes entering the town continues to increase during early morning and reaches a maximal peak around 0700–0730 h. However, the traffic fluxes usually do not diminish until 0900–0930 h which is not shown in the measured concentrations, where a slight decrease can be observed in this period. This is possibly due to an alteration on the traffic profile from the previous hours which is now mainly constituted by gasoline-fueled vehicles, circulating, at first, at low velocity and, afterwards, in intense and compact traffic jams. These traffic jams are certainly the cause for another concentration peaks appearing around 0900–0930 h. Concentrations then decrease until new peaks are observed near lunch hour (1200 h) thus accounting for heavier traffic fluxes, but based on gasoline-fueled vehicles. Concentrations start to rise again during the afternoon, around 1700–1800 h, accounting for traffic leaving the center of the town. Further on, concentrations are still elevated, compared to baseline, until 2100 h, where other, less intense, peaks are reached. After 2100 h, concentrations drop slowly until 2400 h and until 0200–0300 h of the following day. It can be noticed that this daily pattern is reproducible in the next week day.

However, in weekend days, concentrations are much smaller, seldom surpassing the baseline, with scattered peaks during all day, more concentrated near lunch hour and early afternoon. During weekend days there are neither heavy traffic fluxes nor rush hours, which are reflected in the measurements taken. Also during these days, traffic is mainly composed by gasoline-fueled vehicles.

Figure 7 shows the mass concentration variation pattern of PM_{10} measured at the nearby air quality station of Entrecampos, during the same weekdays depicted in Figures 2 and 3: although there are some similarities regarding the occurrence of concentration peaks during rush hours, no sound correlation was found between measured alveolar deposited surface area and PM_{10}, as the involved metric is completely different as explained previously.

Figures 8 and 9 show the measured size distribution measured with the SMPS analyzer: Figure 8 shows Dp,

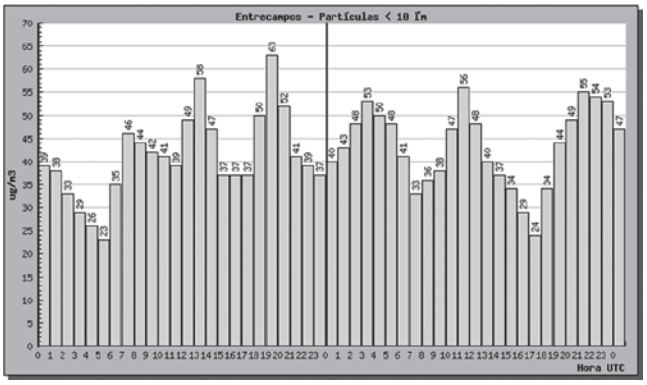

Figure 7. Typical PM10 concentration pattern measured at Entrecampos air quality station during week days: Tuesday, the 10th and Wednesday, the 11th May 2011.

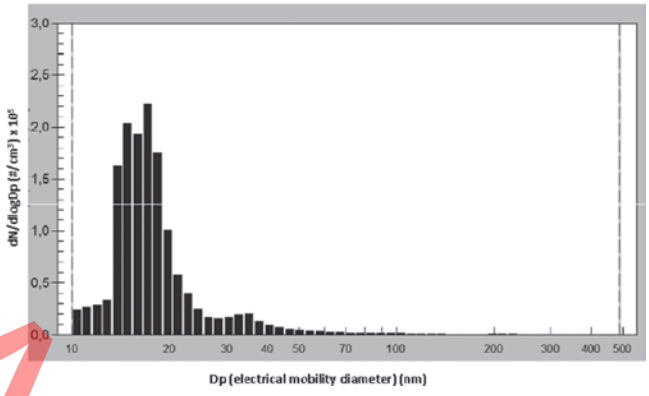

Figure 8. Size distribution of particles during a typical week day: Tuesday, the 10th May 2011.

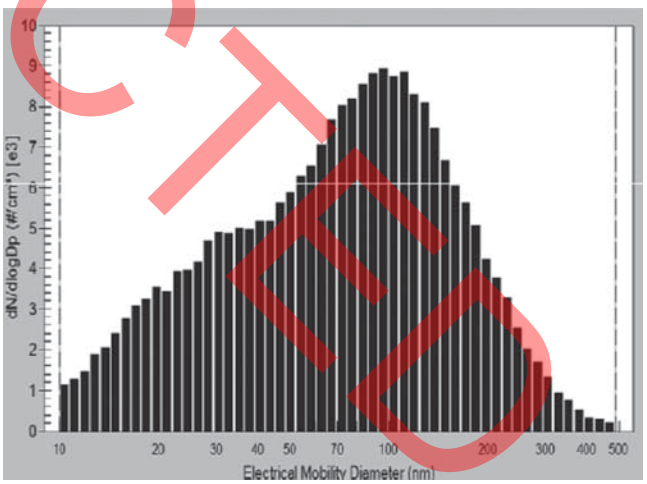

Figure 9. Size distribution of particles during a typical weekend day: Sunday, the 8th May 2011.

the electrical mobility diameter and number of particles on a typical week day, corresponding to an average diameter of 16.9 ± 1.46 nm and an average number of particles (dN/dlogDp) of 4.53×10^4 #/cm³. Figure 9 refers to a typical weekend day, corresponding to an average diameter of 128.6 ± 1.97 nm and an average number of particles of 7.5×10^3 #/cm³. The observed differences, during week

and weekend days, are consistent with a majority of gasoline-fueled vehicles during weekend compared to week days where diesel fueled vehicles (trucks and buses) are in considerable higher number.

Figure 10 shows the evolution of the median values of number of particles and Dp for each day of the week, as computed from the number size distribution spectra measured by the SMPS. Each point is the mean between all days of this study and the error bars represent the respective standard errors of all readings corresponding to each day of the week.

Figure 11 shows a TEM image of ultrafine particulate collected using the nanometer aerosol sampler, during a week day. As expected (Health Effects Institute, 1995) the collected particles seem to be composed of carbonaceous agglomerates of irregular shape, similarly to vehicle exhaust particles from diesel fueled vehicles.

From the values shown in Table 2, it can be noted that outdoor levels are 32% higher during weekend days and 63% higher during weekdays, when compared to baseline levels. Also, it should be noted that the measured deposited surface area is within the same order of magnitude as the values measured by (Kuhlbusch et al., 2004) in the Ruhr area, of 30–45 $\mu m^2/cm^3$ and by (Ntziachristos et al., 2007) in Los Angeles area, of 38–71 $\mu m^2/cm^3$. In what concerns number of particles, (Morawska et al., 2002) measured 6,330/cm^3 on weekends and 8,010/cm^3 on week days for Brisbane; while (Gomez-Moreno et al., 2011) measured 2,000–19,000/cm^3 on week days in Madrid, which means that the measured number of particles in this study is considerably high. In spite of this, the evolution pattern, noticed by (Morawska et al., 2002), on the number of particles and its diameter during week days and weekends was again observed in this study, as shown in Figures 12 and 13.

In what regards the measured levels of deposited alveolar surface area it should be noticed that outdoor levels, during week days, show considerable increases from the baseline, from 160 to 260%. This clearly points out for high exposures to UFP in the urban environment of central Lisbon, as also indicated by the measured total deposited area and lung area covered by particles mentioned

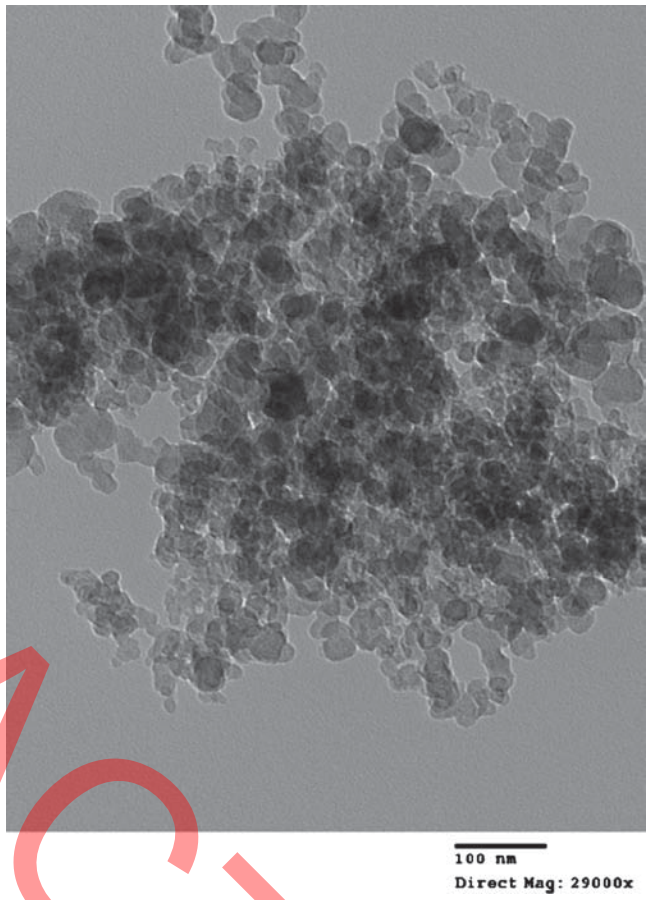

Figure 11. TEM image of ultrafine particulates collected on Tuesday, the 10th May 2011. TEM, transmission electron microscopy.

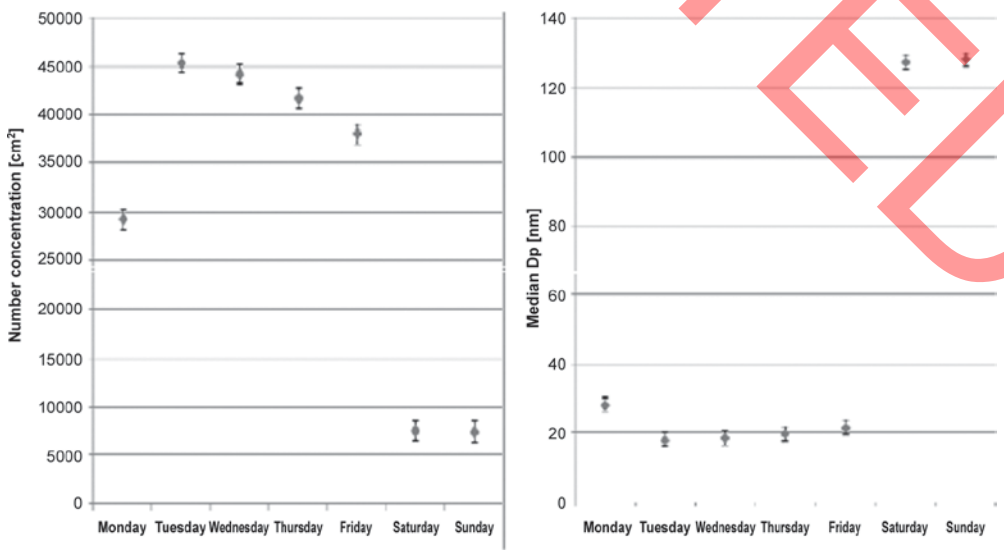

Figure 10. Evolution of size distribution over a typical week: mean particle number concentration and mean particle median electrical mobility diameter as a function of the day of the week based on all the monitoring data obtained during this study, with SE bars on the y-axis.

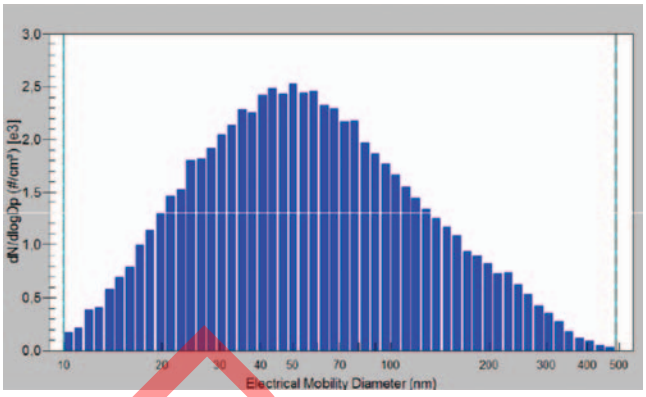

Figure 12. Indoor size distribution of particles during a typical week day.

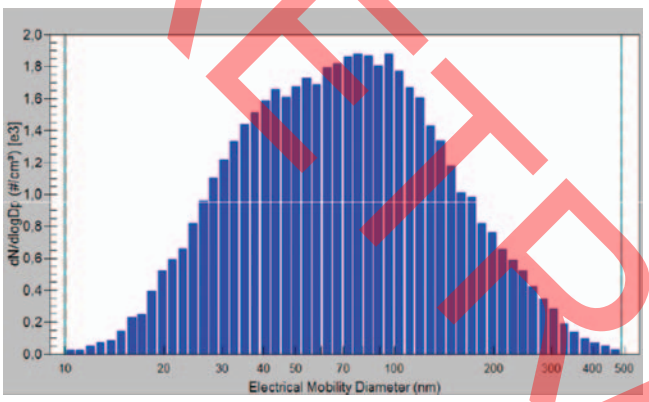

Figure 13. Indoor size distribution of particles during a typical weekend day.

in Table 2, when compared to baseline. This observation is an indicator of a strong potential for occurrence of lung related diseases due to the exposure to high doses of UFP in this urban environment.

Concerning indoor/outdoor ratios, (Kearney et al., 2011) observed that in Windsor, Ontario, outdoor levels were 75–86% higher than indoor levels, which is consistent with the findings of the present study.

When referring to lung-deposited area in TB tract measurements, it was found, as expected (Anastacio & Martin, 2001; Health Effects Institute, 1995; Mauderley, 1992), that both indoor and outdoor levels are lower than the lung deposited area in A tract: comparing values for the same situation, alveolar measurement ranges from 74 to 78% of (alveolar+tracheoronquial) levels. This points out to an increased exposure risk posed by very UFP (which deposit in the alveolar range) in urban environments such as this.

Conclusions

This set of measurements is the first stage of a study comprising the determination of the levels of airborne UFP in the urban atmosphere of Lisbon, with the aim to contribute, further on, to establish epidemiologic correlations on the exposure to exhaust gases.

The study clearly demonstrated the existence of UFP due to automobile traffic which seem to be consistent with observations of UFP concentrations in other major towns. Mainly during week days, observed concentrations can be as high as 2.6 times the measured baseline level.

It should be noted that, although measured parameters such as the deposited area and the lung area covered by particles, are elevated when compared with baseline values, mainly for week days where automobile traffic is more intense, they cannot, at this stage, be ascertained as toxicity indicators. Nevertheless, they point out for the existence of an important contamination of potentially hazardous particles released from automobile traffic in urban environments. Data obtained in this study is basic information for understanding the relationship between exposure to UFP in urban atmospheres and health affections, which can be taken as the basis for epidemiologic studies. As UFP can have a significant lifetime in urban air, possible effects on health cannot be neglected.

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Declaration of interest

The authors declared no conflicts of interest.

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