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Ultrafine particles in ambient air of an urban area: dose implications for elderly

Klara Slezakova[a,b], Jimmy Fonseca[a], Simone Morais[b], Maria do Carmo Pereira[a,*]

[a]LEPABE, Departamento de Engenharia Química, Faculdade de Engenharia, Universidade do Porto, R. Dr. Roberto Frias, 4200-465 Porto, Portugal

[b]REQUIMTE, Instituto Superior de Engenharia do Porto, Instituto Politécnico do Porto, R. Dr. António Bernardino de Almeida 431, 4200–072 Porto, Portugal

*Corresponding author

Maria do Carmo Pereira – phone: +351 22 508 1590; fax: +351 22 508 1449; e-mail: mcsp@fe.up.pt

Klara Slezakova – phone: +351 22 041 4946; fax: +351 22 508 1449; e-mail: slezakok@fe.up.pt

Jimmy Fonseca – phone: +351 22 508 1590; fax: +351 22 508 1449, e-mail: ega08033@fe.up.pt

Simone Morais – phone: +351-22-8340500; fax: +351-22-8321159; e-mail: sbm@isep.ipp.pt

Abstract

Due to their detrimental effects on human health, the scientific interest in ultrafine particles (UFP) has been increasing, but available information is far from comprehensive. Compared to the remaining population, the elderly are potentially highly susceptible to the effects of outdoor air pollution. Thus, this work aims to assess the levels of outdoor pollution at an urban area with emphasis on UFP number concentrations and to estimate the respective dose rates for elderly populations. UFP were continuously measured during three weeks at three sites in north Portugal: two urban (U1 and U2) and one rural used as reference (R1). Meteorological parameters and outdoor pollutants (PM_{10}, O_3, NO and NO_2) were also registered. The dose rates of inhalation exposure to UFP were estimated for three different age categories of elderlies: 64–70 years; 71–80 years; and >81 years. Over the sampling period the levels of PM_{10}, O_3 and NO_2 were in compliance with the European legislation. Mean UFP were 1.7×10^4 and 1.2×10^4 particles cm^{-3} at U1 and U2, respectively, whereas at rural site the levels were 20-70% lower (mean of 1.0×10^4 particles cm^{-3}). Vehicular traffic and local emissions were the main identified sources of UFP at urban sites. In addition, the results of correlation analysis showed that UFP were meteorologically dependent. The exposure dose rates were 1.2–1.4 times higher at urban sites than at reference one, with the highest doses observed for adults with 71–80 years, mainly due to their higher inhalation rates.

INTRODUCTION

Particulate matter (PM) is recognized as one of the most important air pollutants. Up to this date, epidemiological studies have shown association between increase morbidity and mortality rates due to respiratory and cardiovascular diseases and increased levels of ambient PM (Brunekreef et al., 2009; Krewski et al., 2003, 2009; Krewski and Rainham 2007; Samet and Krewski 2007; Turner et al; 2011). The evidence has been so overwhelming that in October 2013 International Agency for Research on Cancer (IARC) classified PM from outdoor pollution as carcinogenic to humans (i.e. Group 1; IARC 2013). In addition to mass and number concentrations, limited number of studies has shown that atmospheric particles of different sizes may be responsible for different levels of adverse effects (Su et al., 2006). The smallest fraction of PM are ultrafine particles (UFP), i.e. those with particle size less than 100 nm (Wang et al., 2011). Unlike larger particles, UFP can cause adverse health effects even at low mass concentrations because of their high number concentrations, high specific surface area, and ability to penetrate into the interstitial spaces of the lungs (Bakand et al.; 2012; Oberdörster et al., 2005;Sioutas et al., 2005). Studies have shown that exposure to UFP are associated with impaired lung function and pulmonary defense mechanisms, inflammatory responses, worsening of respiratory diseases and allergic conditions, cardiovascular problems, and even with carcinogenic and genotoxic consequences (Ferreira et al., 2013; Oberdörster et al., 2001; Stanek et al., 2011). Nevertheless, the mechanisms of UFP health effects are yet to be fully understood. Although epidemiological studies on UFP are needed, exposure assessment issues for UFP are complex (high spatial variability, high seasonal variability in UFP number concentration and composition) and need to be considered before undertaking investigation of UFP health effects (Sioutas et al., 2005).

UFP originate from both natural and anthropogenic sources, being emitted (i.e. primarily origin) as well secondarily formed from gas precursors (Wang et al., 2010). UFP are ubiquitously formed through nucleation (Morawska et al., 2008) and by gas–to–particle reactions and growth processes (including condensation, coagulation and volatilization) (Solomon 2012). However, in urban areas the combustion sources, namely emissions from vehicular traffic are the main sources of UFP (Kumar at al., 2010; Morawska et al; 2008). In addition to the local sources, studies have shown that UFP number concentrations and size distribution are also governed by meteorology, thus creating various patterns (Pirjola et al., 2006; Hussein et al., 2006). In order to fully comprehend these complexities, further studies are needed.

The number of elderly population (i.e. > 65 years) has been increasing throughout the world. Between 1996 and 2008 the elderly population increased from 380 to 500 million (i.e. from 7 to 16% of the total population) (Bentayeb et al., 2012). According to United Nations, in 2050 4% of the world population will be aged over 80 years and 21% will be older than 60 years (United Nations, 2001). For Europe these projections are even higher, with 11% and 29% of the European population being older than 80 and 60 years, respectively (Eurostat 2013). These demographic perspectives bring major consequences for all aspects and areas of human life. Consequently, a better understanding of the health consequences of exposure to various risk factors, notably to environmental ones, including air pollution, are needed, particularly for elderly people. Compared to the rest remaining population, the elderly are potentially highly susceptible to the effects of outdoor air pollution. Nevertheless, the majority of existent studies focuses on other age–population being inexistent the assessment of the exposure to outdoor UFP for elderly.

The present work aims to assess the levels of outdoor pollution at an urban area with emphasis on UFP. The specific objectives of this work was to assess the UFP number concentrations at two urban and one rural site (used as reference) and to estimate the respective dose rates of inhalation exposure to UFP for elderly populations when compared to active adults. The outdoor pollutants (PM_{10}, i.e. particles with aerodynamic diameter below 10 μm, ozone (O_3) and nitrogen oxides (NO and NO_2)) and meteorological parameters (temperature (T), relative humidity (RH), wind speed (WS), precipitation (P), and solar radiation (SR)) were registered in order to characterize the outdoor pollution and weather conditions, as well as, their influence in UFP levels.

MATERIALS AND METHODS

Study area description

Oporto is the second largest city of Portugal, located in the North of Portugal. Its climate is characterized by annual average temperature of 15 ºC approximately and the difference between the highest and lowest monthly averages being less than 10 ºC. Annual air humidity is between 75% and 80%, and the total annual mean precipitation varies between 1000 mm and 1200 mm, with about 40% in the winter season. Prevailing winds are from West and North West (Pereira et al., 2007). The important air pollution sources in the respective area are vehicle traffic, an international shipping port, an oil refinery and a petrochemical complex, a power plant, and an incineration unit (Slezakova et al., 2013).

UFP Collection

UFP were consecutively measured during three weeks of May-June 2013 at three different sites in Portugal. The three sites were selected in order to represent different environments. Sites U1 and U2 were characterized as an urban ones. They were situated in Paranhos district of Oporto city; previously it was demonstrated that vehicular traffic emissions are the main pollution source of this area (Slezakova et al., 2011, 2013). Specifically, both sites were situated within a public garden where senior citizens gathered for social activities (i.e. board-games playing, reading, socializing in outdoor areas of coffee houses, and etc.). The third site R1 was situated in Ermesinde district also in the north of Portugal. This site was considered as a rural background one and was used for comparison. Specifically, R1 was situated in a countryside surrounded by farm plantations and natural forests.

UFP number concentrations in size range 0.02-1 μm were continuously measured daily between 8:30 and 17:30 by condensation particle counters – TSI P-Trak™ (UPC 8525; TSI Inc., MN, USA). Intake flow was 0.7 L min^{-1} and UFP logging interval was 60 s. The samplers were positioned in open area avoiding any obstacles and barriers (trees, bushes walls, and fences) that could interfere with data collection. The equipment were mounted on support (sampling inlets height 1.2 m above the ground) and protected from rain.

Traffic, meteorological and outdoor auxiliary data

The traffic intensity of roads surrounding each site was estimated during two consecutive weekdays. The number of road vehicles was manually counted during every hours between 5:00 and 24:00 h.

Information on outdoor meteorological conditions, namely T, RH, WS, P, and SR were retrieved from the local meteorological station that was located 300–700 m from the sites; all parameters were continuously measured with data registered every 5 minutes. The levels of outdoor pollutants, namely PM_{10}, O_3, NO and NO_2 were provided by Portuguese Environmental Agency. Table 1 summarizes the weather and pollution conditions during the sampling campaigns.

Dose rate exposure analysis

UFP dose rates from inhalation exposure of elderlies were calculated using Equation 1 (Kalaiarasan et al., 2009; Castro et al., 2011):

$$\text{Dose rate (D)} = (BR/BW) \times C \times OF \quad (1)$$

where D is the age-specific dose rate (particle number kg^{-1}); BR is the age-specific breathing rate (L min^{-1}); BW is age-specific body weight (kg); C is the concentration of UFP (number of particles L^{-1}); OF is the occupancy factor (i.e. percentage likely to be in the public garden at a given interval of time). UFP dose rates were estimated for elderlies, i.e. adults > 65 years old. The information on age-specific factors was retrieved from USEPA Exposure Factors Handbook (USEPA, 2011) using BW of 72 kg. BR rates corresponding to sedentary activities (that were the mostly observed) were used as the following: (USEPA, 2011): 4.9 L min^{-1} for seniors 65–70 years old, 5.0 L min^{-1} for seniors 71–80 years old, and 4.9 L min^{-1} for seniors >81 years. OF was considered 2.5 h per day (0.105). For comparison, dose rates of inhalation exposure to UFP were estimated also for active adults (aged 25-64 years) considering the same exposure time (i.e. 2.5 h per day) as for elderlies. Age specific parameters of 4.6 L min^{-1} for BR and BW of 76 kg were used for this group (USEPA, 2011).

Statistical analysis

For the data treatment, the Student's t-test was applied to determine the statistical significance (p<0.05, two tailed) of the differences between the determined means. Spearman's rank correlation coefficient (p<0.05) was calculated to assess the influence of meteorological parameters on UFP number concentrations. All statistical analyses were performed using IBM® SPSS® Statistics software.

RESULTS

Ultrafine particle number concentrations, traffic and meteorological data

The medians and other statistical parameters of UFP at the two urban traffic sites and the rural background site are summarized in Figure 1. The concentrations of UFP ranged between 4.9×10^3 and 4.3×10^4 (mean of $1.7\times10^4 \pm 0.5\times10^4$) at U1 and from 2.4×10^3 and 3.0×10^4 at U2 (mean of $1.2\times10^4 \pm 0.6\times10^4$). At the rural site, the lower levels of UFP were observed with concentrations ranging between 1.5×10^3 and 3.4×10^4 (mean of $1.0\times10^4 \pm 0.7\times10^4$). The statistical analysis of these results indicated that: i) UFP concentrations were significantly higher (p<0.05) at the urban sites than at the rural one; and ii) the differences observed between UFP means at sites U1 and U2 were statistically significant (p<0.05).

The daily profiles of UFP number concentrations at the three sites are shown in Figures 2A–C which also demonstrate the profiles of the traffic density. The average traffic density of the roads around U1 was 16 vehicles min^{-1} and traffic peak hours were detected at 08:30 (24 vehicles min^{-1}) and 17:30 h (25 vehicles min^{-1}). All roads around U1 were characterized by the type of vehicles, mostly constituted by cars (95%). Traffic density around site U2 was comparable with U1 (daily average of 13 vehicles min^{-1}; traffic peak hours at 08:30 and 18:30 with 21 and 19 vehicles min^{-1}, respectively); however, the type of vehicle traffic was different. U2 was situated near a road with a not–negligible proportion of heavy duty vehicles (15%, typically buses). As expected, traffic density at R1 was lower than at the other sites. Small traffic density (< 1 vehicle min^{-1}) was measured at R1 because of the rural location of this site; the road vehicles consisted entirely of passenger cars (100%). The comparisons of UFP number concentration profiles clearly showed that no similarities were observed between rural and urban sites. In addition, the daily profiles of UFP at both urban sites also differed in some extent which suggests different sources and/or influences of UFP at the two characterized urban sites.

The concentrations of UFP at the three sites were also analysed together with the meteorological parameters. Table 2 shows Spearman´s correlation coefficients between UFP number concentrations at sites U1, U2 and R1 and meteorological parameters (temperature, relative humidity, wind speed, and solar radiation). Inverse correlations between the number of UFP, relative humidity and wind speed were observed. Temperature and solar radiation were positively correlated with UFP number concentrations.

UFP dose rates

Dose rates associated with inhalation exposure to UFP that were estimated for three different age categories of elderlies at the three studied sites are shown in Table 3. The results clearly show that: (i) for all age categories the highest dose rates of UFP were found at U1; and ii) for all sites the highest values of UFP dose rates were observed for seniors 71–81 years old.

DISCUSSION

As humans can be adversely affected by exposure to air pollutants in ambient air, European Union has established health–based standards for a number of pollutants in air under the Directive 2008/50/EC. These standards are applied over differing periods of time because the observed health impacts associated with the various pollutants can occur over different exposure times. At this moment there are no air quality guidelines for UFP (Kumar et al., 2011). Still three air pollutants that were monitored in this study are considered in the respective EU legislation, namely particulate matter PM_{10}, nitrogen dioxide and ozone. For ozone, EU sets the legislation standard as a maximum daily 8 h mean with limit value of 120 µg m^{-3}. For nitrogen dioxide the standard is expressed as 1 h mean of 200 µg m^{-3}, allowing 18 exceedances per calendar year. Finally, for PM_{10} the limit value of 24–h average is 50 µg m^{-3} (not being allowed more than 35 exceedances per year) and 40 µg m^{-3} for the annual average. As indicated in Table 1, 24–h concentrations of PM_{10} were lower than 50 µg m^{-3} at all three sites (14 and–30 µg m^{-3} and 10–25 µg m^{-3} at U1 and U2, respectively, and 4–6 µg m^{-3} at R1). Similarly, 1-h measured levels of nitrogen dioxide were lower than EU limits, as well as were the concentrations of ozone. Therefore, over the sampling campaign the levels of the air pollutants were in compliance with the EU legislation.

The concentrations of UFP were significantly higher at two urban sites than at rural ones. Specifically, the UFP levels were, respectively, 70 and 20% higher at U1 and U2 than at R1. Number concentrations of UFP in ambient air can vary by up to five or more orders of magnitude (from 10^2 to 10^7 particles cm^{-3}) depending on environmental conditions and source strengths (Kumar et al., 2010; Solomon 2012). Morawska et al. (2008) reviewed UFP from 71 studies and compared the number concentrations across a wide range of environments, from clean background places to tunnels with levels ranging from 3×10^3 to 2×10^5 particles cm^{-3}. Specifically for urban sites the authors estimated means between 7.2×10^3–10.7×10^3 particles cm^{-3} (based on 24 studies). Additionally, UFP number concentrations at different locations throughout the world were summarized by Wang et al. (2011) who reported concentrations in range of 6×10^3 to 6×10^5 particles cm^{-3}, i.e. in a similar range to those of Morawska et al. (2011). For the European urban sites the latter study reported mean values between 1.2×10^4 (Helsinki, Finland) to 1.9×10^4 particles cm^{-3} (Birmingham, U.K.). It is possible to conclude that the levels of UFP obtained at the two characterized urban sites in Portugal were in the same range as in other European cities. The slight differences (in comparison to those estimated by Morawska et al., 2008) could be caused by the level of urbanization and overall development of area where the sites were located. In addition, seasonal influences, meteorological conditions, different study design (sampling period, duration), and the close proximity of the sampling site to the traffic road at U1 (about 8 m) could account for some of these differences (Seigneur, 2009; Sioutas et al. 2005; Solomon 2012). For rural sites, the information is available only in the study of Morawska et al. (2008) that estimated a mean of 0.48×10^4 particles cm^{-3} (based on 8 studies) which is approximately twice lower than in the present work. Atmospheric formations of UFP, and natural emissions from vegetation (plantations, forests) that were located in the direct vicinity of the site R1 might cause the increased levels (Morawska et al., 2008). In addition, the results in Figure 2C show that no trend between traffic density and UFP number concentrations was observed at site R1 (which was anticipated considering the rural location of this site). However throughout the sampling campaign, soil farming activities (such as soil ploughing) were observed daily during the afternoon hours (approximately from 13:30) at plantations that surrounded the site R1. As demonstrated in Figure 2C, these activities were directly linked with an increase of UFP and may account for some of these increased UFP levels at R1.

Comparing the two urban sites, the daily profiles UFP also differed. At site U1 the daily profile of UFP number concentrations was similar to other urban areas (Solomon et al. 2008; Wang 2011). The peaks of UFP number concentrations and traffic density were observed in the same periods (Figure 2A) indicating that vehicle emissions were the main source of UFP at this site. Vehicle emissions are also a major source of NO_2. At site U1 the typical daily NO_2 trend exhibited concentration peaks during the same hours as UFP (results not shown). Therefore, it is possible to conclude that UFP number concentrations originated mainly from traffic emissions at this site, owing the high levels to the morning and the afternoon traffic rush hours. At U2 (Figure 2B) the trend between UFP and traffic profile was not similar. The first peak of UFP number concentration was observed in in the morning during the rush hour and was associated with motor vehicle emissions. However, the second peak was observed at mid-day–early afternoon (between 12:00–13:00). This increase was associated with emissions of the local soup kitchen that was situated closely to this site U2. Therefore, overall levels UFP at site U2 resulted from contribution of both these sources.

The results of Spearman correlations showed that coefficients between the concentrations of UFP and meteorological parameters, namely temperature, relative humidity, wind speed, and solar radiation were statistically significant (p<0.05) for all variables. The wind speed had a negative correlation with UFP number concentrations due to the greater horizontal dispersion of the pollutants at higher wind speed (Shi et al., 2007). An inverse correlation between UFP and relative humidity can be attributed to the fact that particles can be removed from atmosphere by their dissolution in water droplets (Agudelo-Castañeda et al., 2013) or by the coagulation of droplets on the particles and, thus, be easily removed by below-cloud or in-cloud processes (Wiegand et al., 2011). The positive correlation between UFP number concentration, temperature and solar radiation might be due to photochemical activity, leading to an increase in the concentration of UFP (Park et al., 2008). Specifically, increases in temperature cause an increase of the tropospheric ozone (Elminir 2005). The presence of sunlight then increases photolysis of the troposphere ozone and creates OH radicals that can oxidize precursors. These processes result in the formation of low-volatility species that are able to nucleate under atmospheric conditions (Su et al., 2006; Wang et al., 2010). Overall the obtained findings of the correlation analysis between UFP and meteorological parameters were in agreement with previous studies (Agudelo-Castañeda et al., 2013; Kanawade et al., 2012; Morawska et al., 2008) confirming that formation and levels of UFP in ambient air are meteorologically dependent.

The inhalation exposure dose rates of UFP due to outdoor activities were estimated for three different age categories of elderlies (64–70 years; 71–80 years; and >81 years). At urban sites the exposure dose rates were 1.2–1.4 times higher than at reference one. The highest exposure doses of UFP were found for all age categories at site U1 mostly due to the highest levels of UFP. Evaluating the different age groups, the highest doses of UFP were observed for adults with 71–80 years mainly due to their higher inhalation rate. At this moment there are no other published studies that assessed UFP dose rates of elderlies. In order to better understand the magnitude of UFP exposures, the dose rates of elderlies were compared to those of active adults (25–64 years). The results in Table 3 show that UFP exposure doses rates of elderlies were approximately 15% times higher than those of adults. These results are important because they indicate that elderly might receive higher doses of UFP and thus be at greater risks from air pollution than other age groups. In addition, the elderlies are also more likely to be affected by air pollution, due to generally weaker lungs, heart and defence systems (Bentayeb et al., 2012; Maynard et al., 2003).

The dose rates of UFP estimated in this work were due to outdoor exposure only. However, people spend most of their time (up to 85 %) indoors where they are exposed to UFP from additional sources. The contribution of UFP from outdoors represents approximately only 1–4% (in winter and summer, respectively) of the total UFP daily dose (Buonnano et al. (2014). Therefore, characterization of the respective exposures to UFP for elderly populations in other environments is of upmost importance. The complexity of ultrafine particles though suggests that considerable efforts will be needed in order to properly understand the linkage between the UFP exposures and various types of health outcomes.

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Figure Captions:

FIGURE 1. UFP number concentrations at two urban (U1, U2) and rural (R1) sites: minimum and maximum values, median, 25^{th} and 75^{th} percentile.

FIGURE 2. UFP number concentrations profiles: (A) urban site U1; (B) urban site U2; and (C) rural site R1. The traffic density profile (between 08:00 and 18:00) at each site is also shown.

TABLE 1. Summary of weather conditions (temperature, relative humidity, wind speed, and solar radiation)[a] and outdoor pollution (PM_{10}, O_3, NO and NO_2) during the sampling campaigns at the two urban (U1, U2) and rural (R1) sites. The means are averaged over the 24–h, whereas ranges (in parenthesis) represent ranges of 5–min for meteorological parameters and 1–h means for air pollutants.

	U1	U2	R1
Temperature (° C)	16.6 (15.1–18.7)	13.6 (12.3–16.3)	16.8 (16.1–17.5)
Relative humidity (%)	63 (56–81)	75 (59–90)	89 (84–94)
Wind speed (km h^{-1})	6.3 (4.7–8.5)	6.9 (5.1–10.1)	3.1 (2.3–3.9)
Solar radiation (W m^{-2})	254 (221–269)	312 (278–386)	233 (223–244)
PM_{10} (µg m^{-3})	25 (14–30)[b]	17 (10-25)[b]	5 (4–6)[b]
O_3 (µg m^{-3})	60 (4–111)	59 (12–100)	53 (32–86)
NO (µg m^{-3})	34 (2–224)	15 (2–129)	1.6 (1.3–2.1)
NO_2 (µg m^{-3})	50 (10–134)	29 (8-83)	1.6 (0.5–5.4)

[a]The sampling campaign were conducted in spring period without any rains; therefore the precipitation was 0 mm.

[b]Figures in parenthesis represent concentration ranges of 24-h means during the sampling campaign as settled in EU air quality legislation (Directive 2008/50EC).

TABLE 2.

Spearman correlation coefficients between UFP number concentration and meteorological parameters at the two urban (U1, U2) and rural (R1) sites.

	U1	U2	R1
Temperature (° C)	**0.119**	**0.598**	**0.473**
Relative humidity (%)	**–0.430**	**–0.478**	**–0.630**
Wind speed (km h^{-1})	**–0.136**	**–0.171**	**–0.301**
Precipitation (mm)[a]	–[a]	–[a]	–[a]
Solar radiation (W m^{-2})	***0.108***	**0.178**	**0.581**

Note: values in bold are statistically significant for p<0.01; values in bold italics indicate statistically significance for p<0.05.

[a]Cannot be computed because precipitation was constant (i.e. 0.0 mm) during all period of sampling campaign.

TABLE 3. Estimated dose rates of UFP (particles kg^{-1}) for four different age categories at the two urban (U1, U2) and rural (R1) sites.

		Dose rate (particles kg^{-1})		
		U1	**U2**	**R1**
Elderlies	65-70 years	12.2×10^4 (3.45×10^4 – 3.09×10^5)	8.59×10^4 (1.74×10^4 – 2.15×10^5)	7.25×10^4 (1.07×10^4 – 2.40×10^5)
	71–80 years	12.4×10^4 (3.52×10^4 – 3.15×10^5)	8.77×10^4 1.78×10^4 – 2.19×10^5)	7.40×10^4 (1.09×10^4 – 2.45×10^5)
	>81 years	12.2×10^4 3.45×10^4 – 3.09×10^5)	8.59×10^4 (1.74×10^4 – 2.15×10^5)	7.25×10^4 (1.07×10^4 – 2.40×10^5)
Active adults	25–64 years	10.8×10^4 3.07×10^4 – 2.75×10^5)	7.64×10^4 (1.55×10^4 – 1.91×10^5)	6.45×10^4 0.95×10^4 – 2.13×10^5)

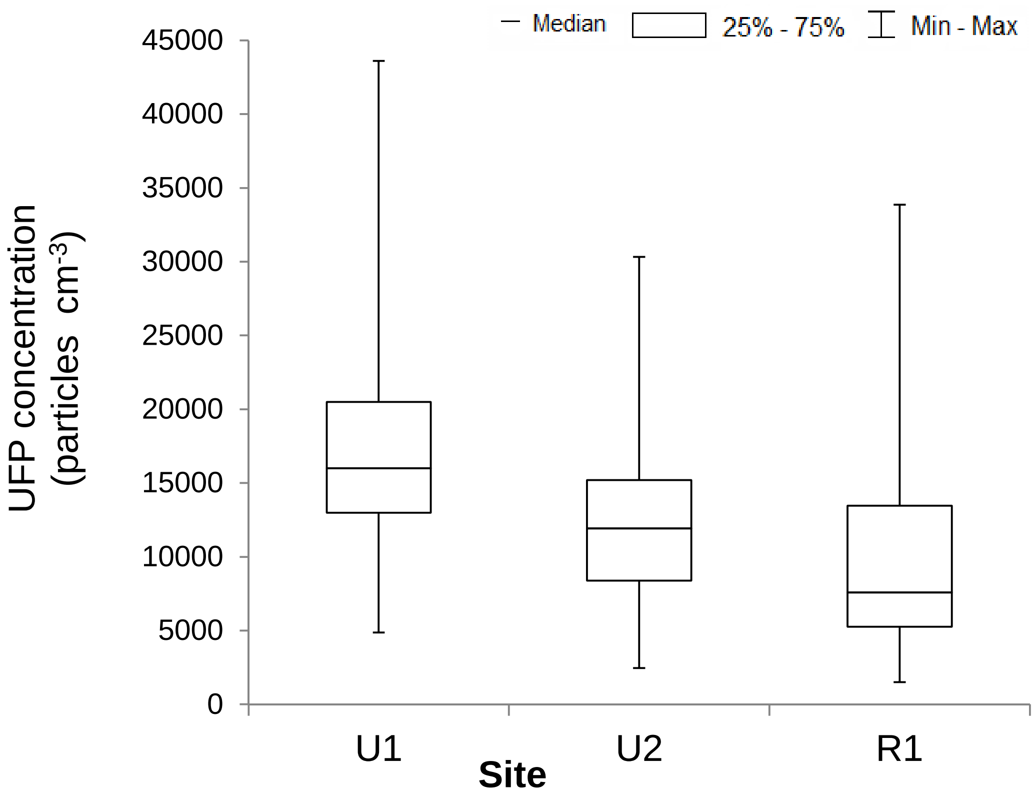
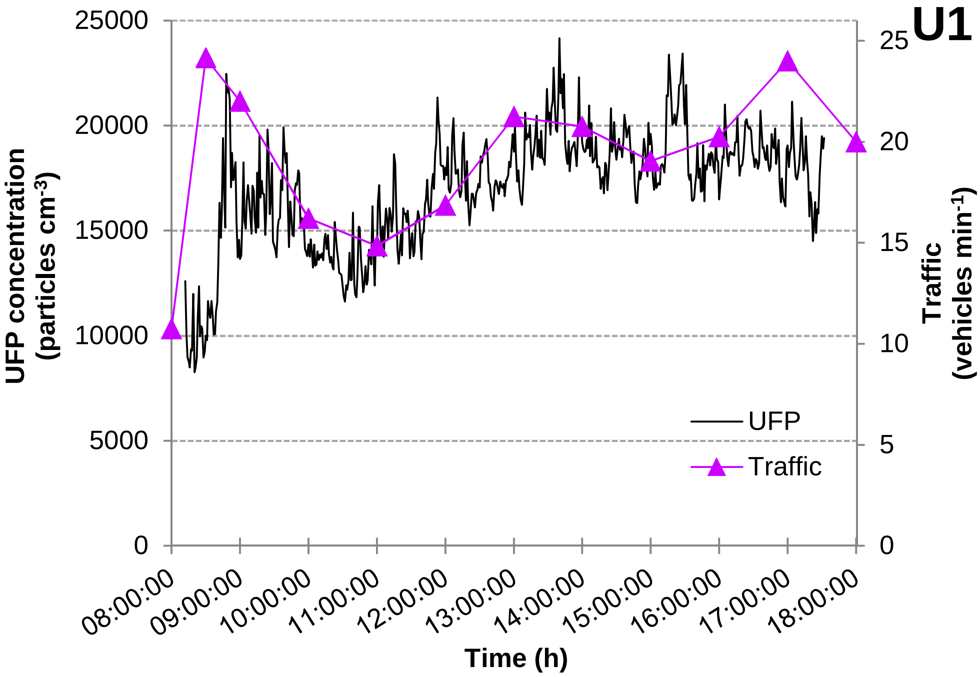
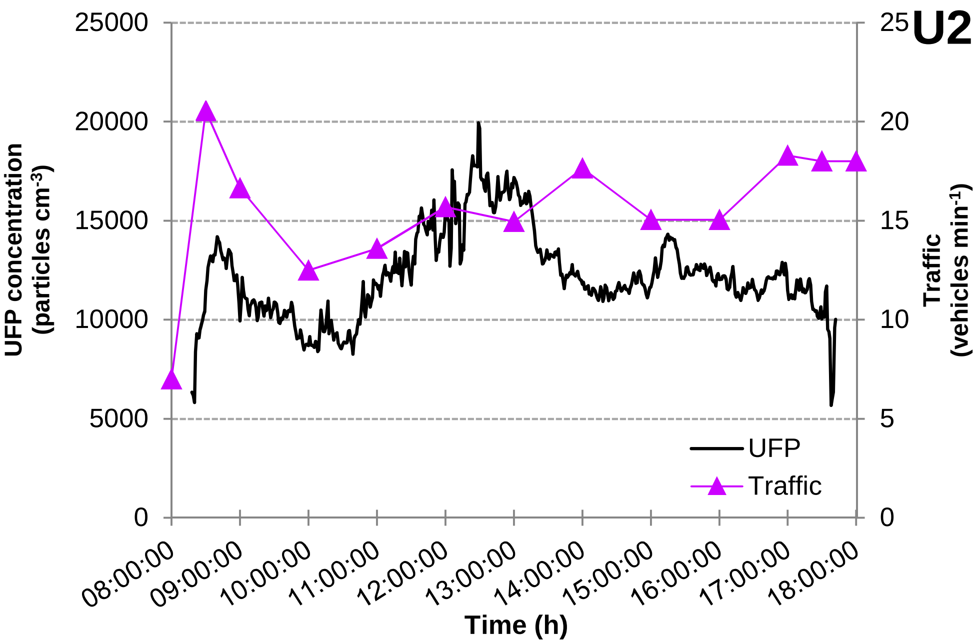

Median
25% - 75%
Min - Max
UFP concentration
(particles cm-3)
45000
40000
35000
30000
25000
20000
15000
10000
5000
0
U1
U2
R1
Site

FIGURE 1

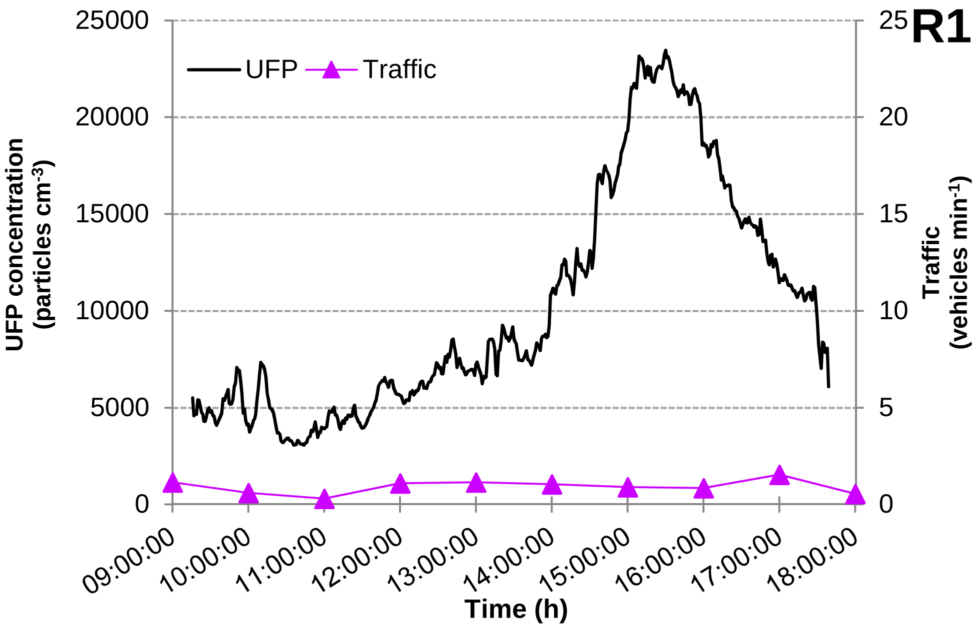
(A)


U1
UFP concentration (particles cm-3)
Traffic (vehicles min-1)
25000
20000
15000
10000
5000
0
25
20
15
10
5
0
UFP
Traffic
08:00:00
09:00:00
10:00:00
11:00:00
12:00:00
13:00:00
14:00:00
15:00:00
16:00:00
17:00:00
18:00:00
Time (h)

(B)


U2
UFP concentration (particles cm-3)
Traffic (vehicles min-1)
25000
20000
15000
10000
5000
0
25
20
15
10
5
0
UFP
Traffic
08:00:00
09:00:00
10:00:00
11:00:00
12:00:00
13:00:00
14:00:00
15:00:00
16:00:00
17:00:00
18:00:00
Time (h)

(C)



R1
UFP
Traffic
UFP concentration
(particles cm-3)
Traffic
(vehicles min-1)
25000
20000
15000
10000
5000
0
25
20
15
10
5
0
09:00:00
10:00:00
11:00:00
12:00:00
13:00:00
14:00:00
15:00:00
16:00:00
17:00:00
18:00:00
Time (h)

FIGURE 2