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ABSTRACT

Conducting aerobic activity on regular basis is recognised as one of the steps to maintain healthier lifestyle. The positive outcomes though can be outweighed if conducted in polluted atmosphere. Furthermore, the specific inhalation during exercising, which results in bypass of nasal filtration systems and deeper penetration into the respiratory system, might result in higher risks especially to pollutants such as ultrafine particles (UFP), which aerodynamic particle diameter are <100 nm. Thus, this work aims to evaluate UFP levels at sites used for conducting physical sport activities outdoors and to estimate the respective inhalation doses considering various scenarios and different physical activities. Monitoring of UFP was conducted during three weeks (May–June 2015) at four different sites (S1–S4) regularly used to conduct physical exercising. The results showed that UFP highly varied (medians $5.1\text{--}20.0 \times 10^3 \text{ \# cm}^{-3}$) across the four sites, with the highest UFP obtained when exercising next to trafficked streets whereas S3 and S4 (a garden and city park) exhibited 2–4 times lower UFP. In view of the obtained UFP concentrations, the estimated inhalation doses ranged $1.73 \times 10^8\text{--}3.81 \times 10^8 \text{ \# kg}^{-1}$ when conducting moderately intense sport activities and $1.93 \times 10^8\text{--}5.95 \times 10^8 \text{ \# kg}^{-1}$ for highly intense ones. Highly intense activities (i.e. running) led to twice higher UFP exposure; children and youths (5–17 yrs old) experienced 203–267% higher doses. Considering the age- and gender- differences, estimated UFP doses of males were 1.1–2.8 times higher than of females. Finally, UFP inhalation doses estimated for walking (commuting to work and/or schools) were 1.6–7.5 times lower than when conducting sport activities. Thus to protect public health and to promote healthy and physically active lifestyle, strategies to minimize the negative impacts of air pollution should be developed and implemented.

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1. Introduction

The society of 21st century enjoys relatively high-quality style with longer life span. However, at the same time, the urbanization and globalization of the modern society and the use of technology have brought unhealthy environmental changes and behaviours. The life style has become more sedentary, which promotes higher prevalence of non-communicable diseases (NCD; WHO, 2015). The last estimates show that about one third of worldwide adult population is insufficiently active (Hallal et al., 2012), which translates to 6% of global deaths (Lee et al., 2012). Furthermore, physical

inactivity combined with unsuitable diet lead to increased obesity (~5% of deaths; WHO, 2010); in EU, 57.4% of the adult population is either overweight or obese, but in some member states this rate is close to 70% (WHO, 2015). Thus, various organizations have been promoting healthy nutrition and more activity lifestyles as a key strategy to improve population health (WHO, 2009; 2015, 2018). Practising aerobic physical exercise can improve overall health (Saunders et al., 2016; WHO, 2018), nevertheless the beneficiary aspects of exercising can be surpassed if conducted in polluted atmosphere (Qin et al., 2019). In EU, the most common settings for conducting physical exercising are parks or outdoors (40%) (EC, 2014). In addition, common physical activities, such as walking and cycling have been increasingly used as means to commute to work and or schools (EC, 2014), which may lead to higher exposure to harmful pollutants. Ultrafine particles (UFP) are typically designated as particles with aerodynamic diameter less than 100 nm

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(HEI, 2013; Kumar et al., 2011, 2013). In the urban atmosphere, UFP are ubiquitous, which implies that humans are always somewhat exposed to them (Holgate, 2017; Morawska, 2017). Combustion is a direct source of UFP in ambient air (*i.e.* various industrial processes, waste incinerations, biomass burning, emissions from naval and air transport; Heal et al., 2012; Paasonen et al., 2013), but vehicle transport is the major source of UFP emissions in urban zones (Carpentieri et al., 2011; Kumar et al., 2014). Secondary formation occurs via photochemical processes and the condensation of semi-volatile vapours (Heal et al., 2012; Morawska et al., 2008). Unlike PM_{2.5}, the physical and chemical properties of UFP undergo constant transformations, which substantially contribute to their large temporal and spatial variability (Kumar et al., 2014). In addition, the current research has indicated that UFP might be more toxic and biologically active than the other PM fractions though the evidence is yet inconclusive (Chen et al., 2016; Lee et al., 2014; Rinaldo et al., 2015; Terzano et al., 2010). Their small particle size allows for deep deposition into the respiratory system (entering the alveoli), from where they may relocate via blood stream and affect various organs and possibly even penetrate the brain (Bakand et al., 2012). Secondly, due to the large surface area, they may carry many harmful chemicals (Chen et al., 2016). Thus, the attention to UFP has been increasing due to their possible adverse health effects (Heal et al., 2012; Heinzerling et al., 2016). While the epidemiological evidence on UFP and health effects is still inconclusive (Clifford et al., 2018; Lanzinger et al., 2016; Stafoggia et al., 2017), exposure to UFP has been linked with hypothesized adverse effects (such as pulmonary and systemic inflammations, oxidative stress, cause-specific, *i.e.* respiratory and cardiovascular, and total mortality; Viitanen et al., 2017; Ohlwein et al., 2019) and various physiological responses (Hertel et al., 2010; Olsen et al., 2014; Stewart et al., 2010; Weichenthal, 2012). Because of the relevance of UFP exposures, various microenvironments have been studied

(such as preschools, primary and elementary schools, age care facilities, homes, offices and occupational settings) (Fonseca et al., 2014; Morawska et al., 2017; Reche et al., 2014; Slezakova et al., 2015, 2019; Viitanen et al., 2017). In regard to exercising, data exist on UFP when being done indoors (Ramos et al., 2014; Slezakova et al., 2018a). However, majority of studies that assessed (or summarized) the impacts of air pollution and physical exercising included pollutants, such as PM₁₀, PM_{2.5}, CO, SO₂, ozone or NO_x (An et al., 2017; Apparicio et al., 2016; Brocherie et al., 2015; Giles and Koehle, 2014; Pasqua et al., 2018; Qin et al., 2019; Slezakova et al., 2018b; Tainio et al., 2016). Information on UFP in the context of physical exercises is scarcer (Endes et al., 2017) but several works assessed exposure to UFP during different modes of commute including physical activities such as biking, cycling, and walking along roads (de Nazelle et al., 2012; Ham et al., 2017; Hofman et al., 2018; Luengo-Oroz and Reis, 2019; Peters et al., 2014; Qiu et al., 2019; Quiros et al., 2013; Rivas et al., 2017).

Thus, this work aims to evaluate UFP levels at sites used for conducting physical activities outdoors. Specifically, UFP were monitored at four different sites that are commonly used for outdoor exercising and the respective inhalation doses were estimated for various scenarios and considering different physical activities.

2. Material and methods

2.1. Study design and UFP measurements

UFP in ambient air were monitored in Oporto Metropolitan Area (Fig. 1), which is the 2nd largest urban area situated in north in Portugal (AMP, 2019). Industrial (an oil refinery and local manufactures) and traffic emissions (road traffic and from shipping) are among the major pollution sources in this area (Slezakova et al., 2011, 2013). The campaigns were conducted during three weeks

Coordinates: 41°9'43.71"N - 8°37'19.03" W

Site position: S1 - S4

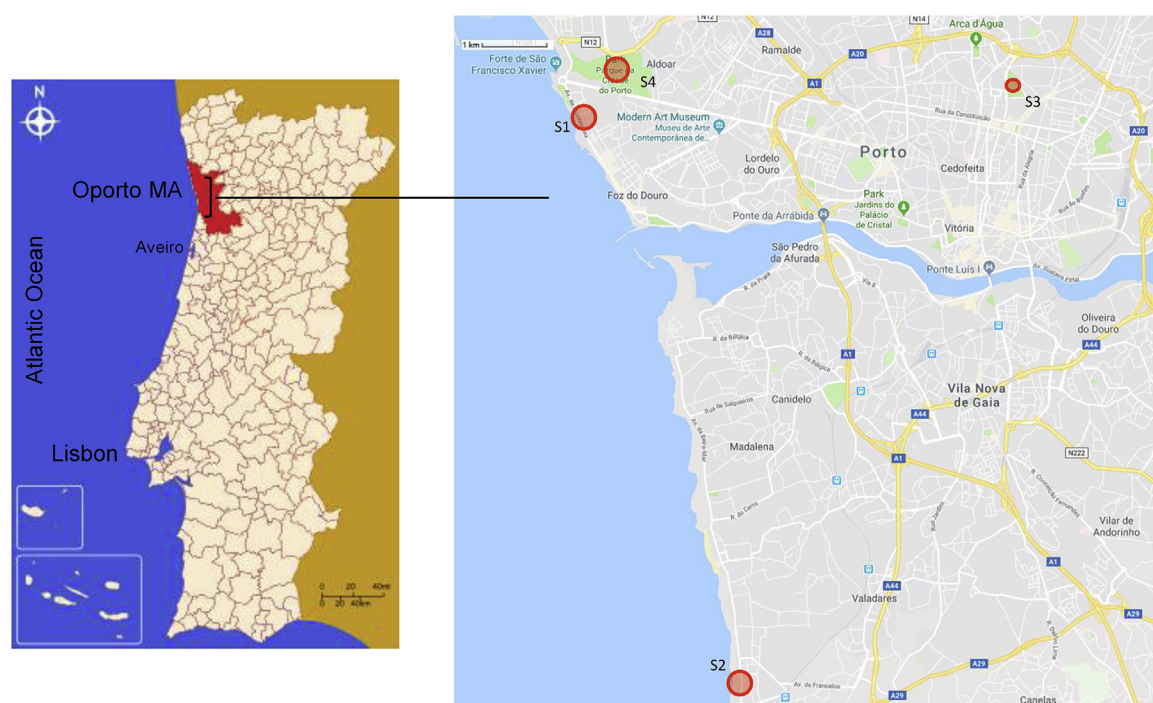


Fig. 1. Locations of four sites to conduct outdoor sport activities (S1–S4) in Oporto Metropolitan Area (Portugal). Note: Red mark indicates the position of each site. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Characterization of the sampling sites S1–S4.

Position	S1	S2	S3	S4
	41° 9' 11.56" N, 8° 40' 43.62" W	41° 4' 38.11" N, 8° 39' 19.93" W	41° 10' 0.39" N, 8° 36' 16.65" W	41° 10' 10.17" N, 8° 40' 27.42" W
Type	Urban	Urban	Urban	Urban
Emissions influences	traffic	traffic	–	background
Traffic density				
Total (range) (vehicles day ⁻¹)	15562 (14004–17754)	9798 (9636–10056)	46 ^a (33–59)	39 ^b (11–79)
Min–Max (vehicles h ⁻¹)	906–1842	600–1116	0–11	0–16
Peaks (vehicle h ⁻¹)	8:00–9:00 1062–1668 17:00–18:00 1668–1842	17:00–18:00 1038–1116	16:00–17:00 6–11	9:00–10:00 1–16
Distance between monitoring point and the closest main road (m)	8 (no obstacles)	8 (no obstacles)	~90 (shielded by vegetation)	~250 (shielded by vegetation and park wall)
Exercising population				
Total (range) (subjects day ⁻¹)	2434 (1800–2340)	1310 (990–1488)	333 (295–388)	2012 (1617–3135)
Min–Max (subjects h ⁻¹)	12–882	20–270	3–110	44–574
Peaks (subjects h ⁻¹)	9:00–10:00 h 162–666 18:00–19:00 h 204–882	9:00–10:00 h 148–270 18:00–19:00 h 58–222	9:00–10:00 h 53–110 18:00–19:00 h 68–82	10:00–11:00 h 208–574 18:00–19:00 h 222–450
General population				
Total (range) (subjects day ⁻¹)	3338 (2016–4140)	269 (200–338)	1457 (1337–1680)	1100 (971–1224)
Min–Max (subjects h ⁻¹)	84–456	1–54	23–336	7–216
Peaks (subjects h ⁻¹)	13:00–14:00 252–456	16:00–17:00 34–54	10:00–11:00 173–257 14:00–15:00 246–336	10:00–11:00 123–151 15:00–16:00 117–216

^a School buses transporting children to the educatory center that was situated in the garden and small trucks used by garden employees.

^b Small trucks used by park employees to service the area.

(May–June 2015) consecutively at four different sites (hereinafter abbreviated as S1–S4), all of which being regularly used to conduct physical exercising. The characteristics of each site are summarized in Table 1 whereas Fig. 1S (a–d) of the Supplementary Material represent the graphical demonstrations. S1 is an urban site with a direct influence of traffic emissions. It is situated on a boulevard in Oporto sea-side where river Douro enters the Atlantic Ocean. It is a very popular spot for exercising (equipped with running line, free drinking-water taps, etc.) as well as for daily walks; it has many outdoor coffees and restaurant esplanades and shops. The boulevard is bordered (right side) by a busy road that (along the river on the left side) connects the sea-side with the city center. Similarly, S2 is situated on a sea-side avenue where physical activities are often conducted (running line along the coast with beaches) but on the southern side of the Oporto Metropolitan Area (Fig. 1), where there was considerably lesser traffic density (Table 1). Monitoring sites S3 and S4 were considered as, respectively, urban and urban background one. They were situated in public enclosed garden (S3) and in the main city park (S4). Both sites were shielded from direct impacts of traffic emissions due to the vegetation, which was evident namely at S4 (Fig. 1 and 1Sd).

At each site, UFP were collected during 3 days, namely Tuesdays to Thursdays (Mondays and Fridays were avoided due to pre-weekend and post-weekend inconsistencies in traffic patterns). Each day monitoring was done during the daylight hours (usually between 7:30 a.m. and 8:30 p.m.) for approximately 12–13 h and when the exercising people were present on sites. UFP were monitored by portable condensation particle counters (P-Trak model 8525, TSI Inc., MN, USA) that detect the size of particles between 20 and 1000 nm (in range of $0-5 \times 10^5 \# \text{ cm}^{-3}$; TSI, 2013). The continuous monitoring was done with logging interval of 1 min (i.e. continuous measurements with an average over 60 s being recorded) and intake flow of 0.7 L min^{-1} . In total, 2335–2460 measurements were register at each site. The equipment was placed on supports (shielded from a rain and/or direct sun exposure) approximately 150 cm above the ground and positioned away

from any obstacle or physical barriers (vegetation, fences, walls, etc.), which might hamper the data collection. Prior to the UFP monitoring, all the instrument was cleaned and calibrated (at manufacturer) and per their recommendations zero readings were daily checked in order to confirm the normal operations. In order to verify data acquisition differences between the instruments, validation tests were conducted with no statistically significant differences found. The alcohol refill (100% isopropyl) was performed every 6–7 h by a researcher who was constantly present at the site. This procedure implied 2–3 min interruptions during the data collection. Information on meteorological conditions during the respective consecutive periods of campaign was also registered (Table 1S). Concentrations of ambient air pollutants (namely PM₁₀, PM_{2.5}, NO, NO₂, NO_x, O₃; Table 2S) were retrieved from the Portuguese air quality monitoring network (PEA, 2019) for each site from a station that was the closest (maximal distance < 1.5 km). These data were then treated and used as a proxy of the outdoor conditions at each site. Traffic density was obtained by manual counts (Fig. 2S) and a detailed record about happenings at each site was daily provided with any unusual occurrence (that might influence the data collection) registered. Finally, the number of exercising people and detailed description of their specific activities were also documented (Figs. 3S–4S).

2.2. UFP dose calculations

The methodology for calculation of inhalation dose (D) was described previously (Slezakova et al., 2018a, 2019) and was calculated as:

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

where D is the age-specific dose ($\mu\text{g kg}^{-1}$); BR and BW represent the age-specific breathing rate ($\text{m}^3 \text{ min}^{-1}$) and body weight (kg), respectively; C is the median concentration of the UFP pollutant across all sampled days ($\# \text{ m}^{-3}$); and t is time of exposure (min). For

the reader's convenience, further details of dose assessment are summarized in Supplementary material (Text 1S) whereas an example of a calculation is given in Table 3S. The exposure scenarios were based on WHO global recommendations for additional health benefits (WHO, 2010) which imply that: (i) children and youth (5–17 yrs) should perform daily physical activity greater than 60 min of moderate-to-vigorous-intensity; (ii) adults (18–64 yrs) should engage in 300 min per week of moderate-intensity aerobic physical activity or 150 min per week (about 1 h per day on six days per week basis) of vigorous-intensity aerobic physical activity; and (iii) adults (>65 yrs) should follow the general guidelines (i.e. 150 and/or 300 min of moderate or vigorous activity, respectively) unless specific medical conditions indicate the contrary. Additional information regarding the relevant gender- and age-specific parameters (USEPA, 2011) are also provided in Text 1S.

2.3. Statistical analysis

The statistical data treatment was performed using the Microsoft Excel 2013 (Microsoft Corporation), SPSS (IBM SPSS Statistics 20) and Statistica software (v. 7, StatSoft Inc., USA). In order to assess if the sample data followed a normal distribution Shapiro–Wilk's tests were conducted. Once no site presented a skewness higher than $|3|$ ($0.332 > s < 2.42$) nor a kurtosis higher than $|10|$ ($-0.330 > k < 6.25$), it may be assumed that no severe deviations from a normal distribution (Fig. 5S) were present (Kline, 2005). The obtained data were normalized using the z-cores and one-way ANOVA was used to compare the differences between the obtained means. Nonparametric Mann–Whitney *U* test was also used for comparison of the obtained medians. Statistical significance was set as $p < 0.05$. Multivariate regression model was used to investigate the simultaneous relationship between ultrafine particle number concentrations, meteorological parameters and traffic.

3. Results

UFP measured at all sites are summarized in Fig. 2 whereas the respective values are summarized in Table 4S. The one-way ANOVA results ($F(3, 2333) = 1102.80$) showed that the obtained means were significantly different between all sites (all $p < 0.001$). Overall, UFP ranged with absolute minima and maxima reaching values of $1.1 \times 10^3 \text{ \# cm}^{-3}$ (at S4) and $51.4 \times 10^3 \text{ \# cm}^{-3}$ (at S1). The highest UFP median ($20.0 \times 10^3 \text{ \# cm}^{-3}$) was observed at S1 being twice higher than at S2 (median of $8.7 \times 10^3 \text{ \# cm}^{-3}$).

Diurnal patterns of UFP at four sites are presented in Fig. 6S, whereas Fig. 3 shows time series analysis of UFP (30 min average concentrations) at S1–S4 as well as the mean vehicles counts during each hour at each site. These results show that at S1 and S2 morning traffic rush hours (~8:00–9:00) corresponded to peaks in particle number concentrations. A similar trend was also observed at S1–S2 during afternoon traffic rush hours (UFP peak at around 17:30–18:30). This increase was especially noticeable at S1 where diurnal trends of UFP (Fig. 6S) showed approximately 50–60% higher concentrations in later afternoon hours (16–20 h) than in the morning (8–10 h) or midday period (10–14 h). At this site, UFP increased (at approximately 10:30 a.m.) and continued to rise during afternoon hours until the end of day (Fig. 3); this pattern was registered on various days of sampling. At both S1 and S2, hourly means of UFP were significantly and positively correlated with the number of vehicles (Spearman correlation coefficient (r_s) = 0.770 and r_s = 0.659 at $p < 0.01$). On the contrary, at S3 and S4 hourly UFP means were not associated with vehicles counts (r_s = 0.064 and r_s = 0.197). Finally at all S1–S4, another rise of UFP number concentrations occurred at the middays (during which traffic counts were declining).

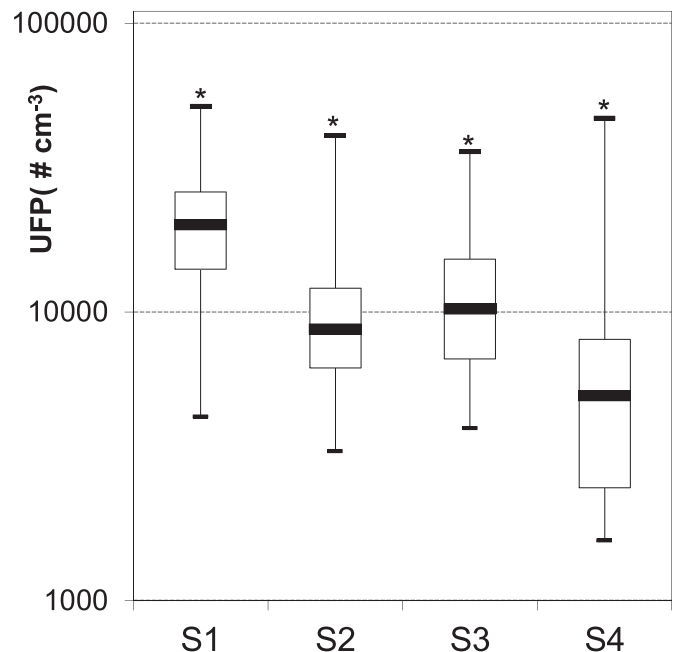


Fig. 2. Ultrafine particles (UFP) levels (■ median; □ 25–75%, and ± range) at four characterized sites S1–S4. Note: across all sites, the medians UFP were significantly different ($p < 0.05$).

The analysis of UFP number concentrations at S1–S4 together with the meteorological parameters (Table 5S), namely air temperature, relative humidity, wind speed, and solar radiation showed that the obtained Spearman correlation coefficients (r_s) between the UFP and the meteorological parameters were statistically significant ($p < 0.01$) for all variables, but in general demonstrated low strength of associations (except for solar radiation). At all sites, UFP concentrations were positively correlated with temperature, whereas negative relationships between UFP and wind speed, as well as relative humidity were detected. However, the simultaneous analysis (by multivariate regression model) between UFP, meteorological parameters (temperature, relative humidity, wind speed and solar radiation) and traffic density showed low associations (Table 6S), with R^2 ranging between 0.471 at S4 and 0.578 at S2, respectively. Solar radiation was the significant parameter ($p < 0.05$) for UFP levels at S4, whereas solar radiation and wind speed were significant at S3; temperature, and relative humidity and temperature were the significant parameters for UFP at sites S2 and S1, respectively.

Fig. 4a shows the doses estimated due to inhalation to UFP during sport activity (based on WHO recommendation scenarios) at S1–S4. In this context, moderate physical activities included bicycle riding, skating or power walking, whereas vigorous activities were those such as running and football playing. Across S1–S4, UFP doses largely varied. Considering the indicated age groups, the medians of inhalation doses due to these activities (across four sites) were between 1.73×10^8 and $3.81 \times 10^8 \text{ \# kg}^{-1}$ for moderate activities, and 1.93×10^8 – $5.95 \times 10^8 \text{ \# kg}^{-1}$ for the vigorous ones. The highest ones (2.85×10^8 – $1.59 \times 10^9 \text{ \# kg}^{-1}$) were associated with S1 (~2–4 times higher than at S2–S4), which was the most polluted place in terms of UFP. Evaluating the activity, the highest estimated doses of UFP were observed for intense sport activities (i.e. running) with the inhalation dosage range of 7.28×10^7 (>64 years old at S4)– $1.04 \times 10^9 \text{ \# kg}^{-1}$ (5–17 yrs at S1). Concerning the different age groups, the highest inhalation doses were found at all sites for children and youth (range of $1.19 \times 10^8 \text{ \# kg}^{-1}$ at S4 and

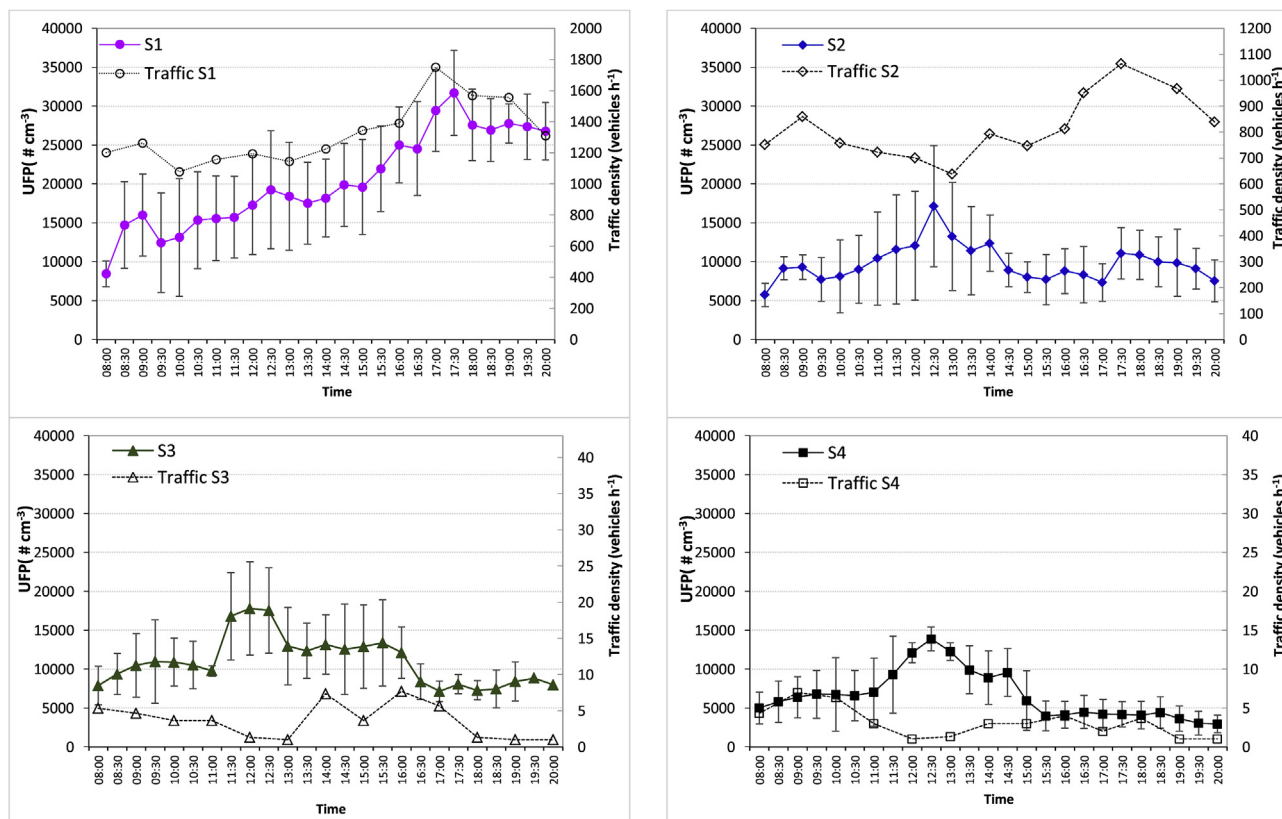


Fig. 3. Time series variation of average ultrafine particle (UFP) concentrations at four sites (S1–S4). Note: Due to the high resolution of the measurements (1 min), the graphic representations correspond to 30 min means (averaged over the entire period of measurements); for better illustrations of error bars (represent standard deviations), the plots are shown individually for each site. The dashed lines represent average traffic profiles (vehicles numbers, by hourly counts) in the streets surrounding S1–S4 (please note different scales on secondary vertical axis y to better visualization of the patterns).

$4.65 \times 10^8 \text{ \# kg}^{-1}$ at S1 during moderate-intensity exercising; $2.65\text{--}11.9 \times 10^8 \text{ \# kg}^{-1}$ for highly-intensive activities).

Fig. 4b demonstrates the inhalation doses to UFP estimated for adult population (>17 yrs) for females vs. males exercising at S1–S4. The comparison between both genders showed that estimated UFP doses of females were 1.1–2.8 times lower than of male, with the highest differences observed in the younger populations (151–164% for 18 to <21 and 21 to <30 yrs old). UFP inhalation dosage estimated for a general population (Fig. 4c), considering 30 min and 1 h of walking, resulted in dose 1.6–7.5 times lower than when conducting the sport activities with the highest dose dosage estimated at S1 ($8.27 \times 10^7\text{--}2.75 \times 10^8 \text{ \# kg}^{-1}$) at S1.

Dose scenarios estimated for the periods with the highest number of exercising subjects (afternoon: ~7 p.m. at all sites; morning: ~9 a.m. at S1 and S3, and ~10 a.m. at S2 and S4) using average of UFP of each hour are demonstrated in Fig. 5. Overall the highest doses were received by subjects exercising at S1 during afternoon hours ($3.89 \times 10^8\text{--}1.42 \times 10^9 \text{ \# kg}^{-1}$), being 40–45% higher than those at morning at the same site and 50 (S2) to 600% higher than at other sites.

4. Discussion

The obtained UFP concentrations in this work (Fig. 2, Table 4S) were in general agreement with the previous research that has emphasized the large intra-city spatial variations of UFP (Li et al., 2018, 2019). Buonanno et al. (2011) reported spatial differences in mean UFP varying by factor of two (Cassino, Italy), whereas Saha et al. (2019) observed factor of three in variability of mean urban

UFP concentrations measured across 32 sites in Pittsburgh (USA) or even by factor of nine in Dresden, Germany (Birmili et al., 2013). In addition, various works reported a rapid decline in UFP concentrations with increasing distance from the source, i.e. from road (Fujitani et al., 2012; Karner et al., 2010; Kumar et al., 2014; Pattinson et al., 2014). Though at both S1 and S2 the distance from the road was the same (8 m), S1 was the site with the higher traffic density (Table 1; median of 1311 vehicles h^{-1} vs. 793 vehicles h^{-1} at S2). However, it needs to be further emphasized that UFP number concentrations might be somewhat underestimated as particles below <20 nm (lower limited if used samplers) were not detected in the current work. While previous research has demonstrated the importance of this small fraction mostly in atmospheric processes, especially in the formation of ultrafine particles (Kirkby et al., 2016; Kulmala et al., 2007), more recent data have highlighted traffic and, more specifically, vehicle exhausts as relevant sources of particles in diameter range of 1.3–3.0 nm to urban air (Rönkkö et al., 2017). S3 and S4 were situated in much greater distances from the main roads (Table 1). Apart from the distance and lesser amount of vehicles in the direct vicinity of the monitoring points (<4 and <3 vehicles h^{-1} , respectively), the existent traffic was different. Trucks and buses consisted 22–50% at S3 and 60–99% at S4 vs. 2–6% at S1 and S2. In comparison to S1, S3 exhibited twice lower UFP levels (median of $10.2 \times 10^3 \text{ \# cm}^{-3}$) whereas the lowest UFP were observed at S4 ($5.1 \times 10^3 \text{ \# cm}^{-3}$).

As there are no legislative limit or guidelines for UFP in ambient air (Kumar et al., 2011), it is rather difficult to compare the existent levels from a health-related perspective. However, because of the importance of UFP, the number of studies have been emerging. In

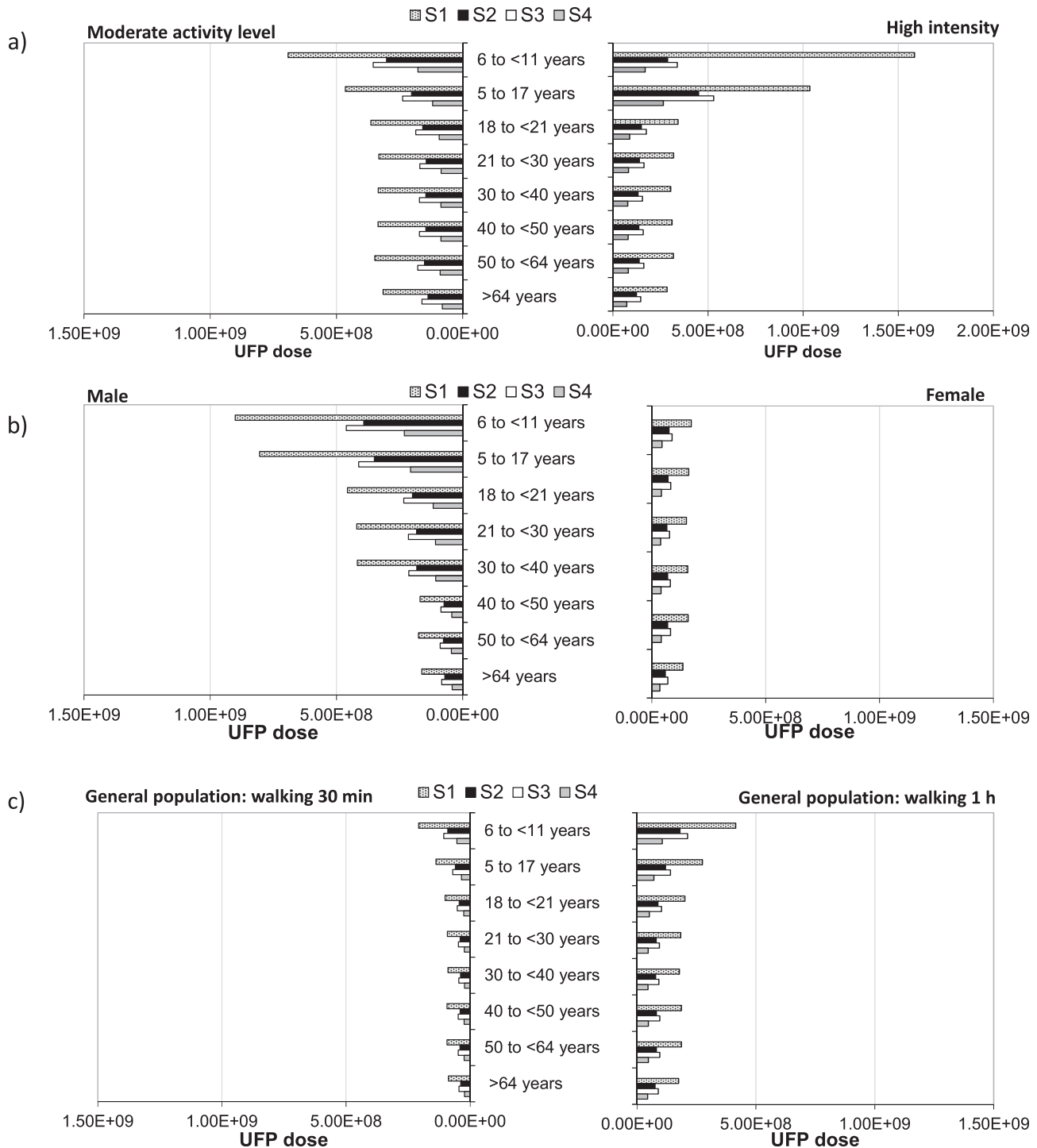


Fig. 4. Inhalation dose due to exposure to ultrafine particles (UFP) during different physical activities at four sites S1–S4: (a) based on WHO recommendations (WHO, 2018) of 300 or 150 min per week of moderate-intensity or vigorous-intensity physical activity, respectively; (b) based on gender-, age, frequency and duration specific parameters (INE, 2016); (c) for general public while walking on the streets. Note: young children and youths (5–17 yrs old) are not included in the gender- and activity-specific scenario (b) due to the data unavailability for this age group.

terms of concentration ranges of ambient UFP, one of the first and still most comprehensive studies was reported by Morawska et al. (2008) who summarized the existent concentrations of UFP based on different characteristics of the sites. Considering eight different types (ranging from clean background sites to on-road sites and tunnels; Table 7S), the authors (Morawska et al., 2008) reviewed all the available studies (72) and estimated the respective UFP ranges.

For urban sites (as to similar to S1–S3 of this work), the reported mean was $10.76 \times 10^3 \text{ \# cm}^{-3}$ (median of $8.83 \times 10^3 \text{ \# cm}^{-3}$), whereas for urban background sites (corresponding in this study to S4), mean of $7.29 \times 10^3 \text{ \# cm}^{-3}$ (median of $8.10 \times 10^3 \text{ \# cm}^{-3}$) was estimated (Morawska et al., 2008). The values in this work resembled those estimations, namely for sites S2–S4 as the obtained UFP means were 10.0×10^3 and $11.6 \times 10^3 \text{ \# cm}^{-3}$ at S2 and

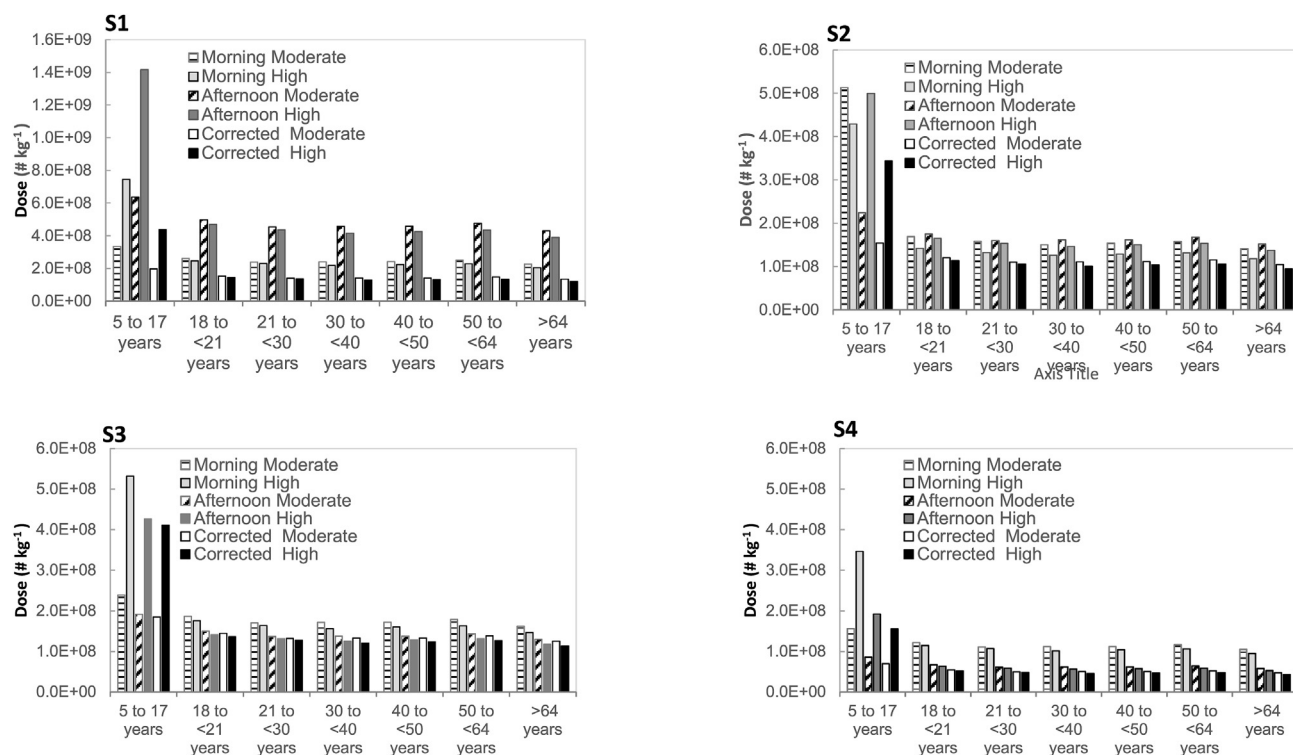


Fig. 5. Inhalation dose to ultrafine particles (UFP) at four sites S1–S4 during different periods of day. Note: duration of moderate-intensity and vigorous-intensity physical activity, respectively, correspond to WHO recommendations (WHO, 2018) of 43 or 22 min per day of moderate-intensity or vigorous-intensity physical activity, respectively. Morning and afternoon UFP dose corresponds to hours with the highest number of exercising subjects (afternoon: 7 p.m. at S1–S4; morning: 9 a.m. at S1 and S3, and 10 a.m. at S2 and S4). Corrected dose corresponds to hours with lowest UFP concentrations (mornings: before 8 a.m.; evening: ~20:00).

S3, respectively, and $7.1 \times 10^3 \# \text{ cm}^{-3}$ at S4. At S1, the obtained UFP were twice higher (mean of $20.3 \times 10^3 \# \text{ cm}^{-3}$) than those estimated. To provide some level of comparison, considering other southern EU countries, the reported levels obtained in this work were similar to those observed at traffic emissions influenced sites in Athens ($27 \times 10^3 \# \text{ cm}^{-3}$ for N_{7-3000} ; Puustinen et al., 2007). In other relevant cities, such as Rome or Barcelona, reported UFP were much higher (N_{10} : $46.8 \times 10^3 \# \text{ cm}^{-3}$ and $59.2 \times 10^3 \# \text{ cm}^{-3}$, respectively; Paatero et al., 2005). Obviously, the latter two are much larger and busier metropolitan areas and apart from different levels of urbanizations and meteorological conditions that are relevant for particle concentrations (Meier et al., 2015), both studies implied different study protocols (monitoring approach and equipment) and measured different number fraction of UFP (in terms of lower cut-point). More recently, within the BREATHE study conducted in Barcelona (Reche et al., 2014; Rivas et al., 2014, 2015), UFP (N_{10-700}) mean of $23.4 \times 10^3 \# \text{ cm}^{-3}$ (vs. $20.3 \times 10^3 \# \text{ cm}^{-3}$ at S1) was reported across 36 sites, and similarly to this work, with UFP levels approximately 40% higher at sites with traffic emissions; whilst for other urban cities (Birmingham, UK) some similarity with UFP at S1 also exist (Birmingham, UK: $19 \times 10^3 \# \text{ cm}^{-3}$; Wang et al., 2011).

Concerning the correlation between UFP and meteorological parameters, the negative associations with wind speed were most likely caused by greater dispersion of UFP at higher wind speed (Shi et al., 2007), which might have influenced UFP concentrations profiles and have caused temporal variations of the respective levels. Particle removal from the atmosphere (either by the coagulation of droplets and/or by dissolution in water droplets) and then consecutive cloud processes (Agudelo-Castañeda et al., 2013; Wiegand et al., 2011) might have caused the inverse associations with relative humidity. Regarding temperature, the positive

correlations was likely a result of the atmospheric photochemistry (Kanawade et al., 2012; Park et al., 2008). Nevertheless, the respective correlations (though being significant) were relatively weak, which might be associated with UFP distributions; the predominant impacts of temperature were reported mainly for the lower particle size ranges (N_{15-50}) (Jamriska et al., 2008; Wang et al., 2010). Thus, in the future studies, it would be relevant to investigate the specific the distribution of UFP and the impacts for different particle size fractions. Furthermore, solar radiation (unlike other meteorological parameters) presented moderately–strong correlations with UFP ($r_s = 0.591\text{--}0.622$) at all four sites. Similarly, several studies reported associations between increased UFP, solar radiation, ozone-initiated processes and photochemistry (Agudelo-Castañeda et al., 2013; Bekö et al., 2015; Brines et al., 2015; Wang et al., 2010, 2011), as sunlight increases photolysis of the tropospheric ozone. Ozone was significantly correlated with UFP at all four sites showing moderately strong associations ($|r_s| = 0.492\text{--}0.587$). Others pollutants did not present significant associations, with exception to S1 where UFP were positively with other traffic pollutants, namely $\text{PM}_{2.5}$ ($r_s = 0.651$) and NO_2 ($r_s = 0.751$), most likely due the same origin (i.e. traffic emissions) of these three pollutants.

Whereas at both S1 and S2 UFP were correlated with number of vehicles (Fig. 3), the contributions of other emission sources cannot be excluded. At S1, possible impacts can be due to local cafes and restaurants, previously other works reported similar impacts on urban UFP (Peters et al., 2014; Saha et al., 2019). It shall be remarked that UFP are more spatially heterogeneous than other regulated pollutants (i.e., $\text{PM}_{2.5}$, NO_2 ; Saha et al., 2019), whereas other works highlighted the large temporal (day to day and seasonal) variations (Peters et al., 2014; de Jesus et al., 2019; Li et al., 2018). The limited number of sites and the consecutive regime of UFP monitoring

during one season were among the primary concerns for this study. Hence, future studies should include larger dataset with longer periods and parallel UFP monitoring at the various sites. In addition, studies with detailed investigation of exposure patterns to UFP are needed. UFP means were not associated with vehicles counts at S3 and S4. At these two sites, UFP levels during the midday periods (around 10–14 h; Fig. 6S) were 10% (at S3) to 150% (S4) higher than those of mornings and late afternoons, respectively. It is also evident that UFP concentrations profiles and traffic patterns showed clearly different trends at these sites (Fig. 3). Finally, the midday rise of UFP (especially noticeable at S2–S4; Fig. 3) was in agreement with the previous studies that also reported elevated midday UFP due to association with nucleation processes mediated by photochemistry (Reche et al., 2014; Kumar et al., 2014; Wang et al., 2010). Nevertheless, these findings should be further confirmed. Particle size distribution and chemical composition would be the key parameters for better identification of UFP origin and emission sources in the respective urban area (de Jesus et al., 2019).

The previous research has shown that conducting outdoor sport activity in places with heavier air pollution might become harmful to health (Kubesch et al., 2015; Qin et al., 2019; Sinharay et al., 2018). Pasqua et al. (2018) reported up to 66 times higher exposure to airborne particulates and no additional health benefits after 15 min of the activity if exercising in highly polluted cities; after 75 min of sport activity the positive impacts of exercising were completely suppressed. In this work S1 (the one with the highest estimated doses) was the most frequented site to exercises (up to 8 times more subjects; Table 1) thus showing the relevance of the respective spot and its air quality for public health implications. On the contrary, estimated UFP dose were the lowest when exercising in a city park (S4) that was secluded from the surroundings. Up today, the majority of the studies concluded that when practicing outdoor sport activities in low polluted environment, the benefits of exercising will by far exceed the adverse effects of air pollution (Giles and Koehle, 2014; Qin et al., 2019). Thus while air quality indices should be developed to better inform sport practitioners where to engage in physical activities, in the meantime simpler guidelines (such as avoiding trafficked streets, preference of sites with vegetation and trees, avoiding rush hours for exercising) should be promoted in order to minimize the negative effect of air pollution on health when exercising.

The highest estimated UFP doses were observed for intense sport activities. It is necessary to point out that under the WHO recommendation scenarios (WHO, 2018), vigorous sport activities were conducted by half less times than the moderately-intense ones (i.e. 150 vs. 300 min per week). Nevertheless, the respective doses were still up to 2.4 times higher than for moderate activities. As ventilation rate and breathing frequency are highly elevated during the intense physical activity, they may cause much larger air inhalation exposure (Londahl et al., 2007; Qin et al., 2019). When exercising, the proportion of UFP that remains deposited in the airways is elevated (0.83 during exercise vs. 0.65 at rest for healthy adult subjects; Chalupa et al., 2004; Daigle et al., 2003). The combined effect of increased ventilation rate and higher UFP deposition may then lead up to a 6– to 10-fold increase in particle number deposited in the airways while exercising (Giles and Koehle, 2014; Oravijärvi et al., 2011). It is noteworthy that highly-intense physical activities were predominantly conducted at S1 (~40% running; Fig. 3S), where UFP levels were already elevated in comparison with other sites further increasing the respective dose and the associated risks. Moreover, when exercising with high intensity (i.e. ventilation rate at 35 L min⁻¹), inhalation changes from a nasal to a mouth-predominance (Brocherie et al., 2015; Wagner and

Clark, 2018) and the bypass of the nasal filtration systems might further increase the pollutant dose (Giles and Koehle, 2014). In addition, several studies have been highlighting that UFP exposure is not directly proportional to number concentrations as UFP deposition within respiratory system is size-dependent (Kumar et al., 2014); thus information on particle size distributions would be important for the accurate estimation of dose rates.

UFP doses estimated for youth and children were the highest ones (Fig. 4a). Firstly, the time under this scenario was longer for children and youth since WHO suggests a minimum of 60 min per day (vs. 150 or 300 min per week for adults; WHO, 2018). Secondly, due to their higher minute ventilation children (whilst their physiological and immunological systems are still developing) receive a higher dose of airborne particles relative to lung size compared to adults similarly to the previously reported (Burtcher and Schüepp, 2012; Morawska et al., 2013; Laiman et al., 2014). From the physiological point of view, the body weights and inhalation rates of young children are different from those of youth. The USEPA reports 21 kg and 10 m³ day⁻¹ for 4–6 years old children whereas values are 67 kg and 16.3 m³ day⁻¹ for adolescents 15–19 years old (USEPA, 2011); data for specifically Portuguese population are not available. To oblige the WHO scenarios (age category 5–17 yrs), in this work the respective results were calculated using the medians across those ages (USEPA, 2011). The respective conclusions need to be considered carefully. Furthermore, susceptibility of the individuals was not accounted in this work.

Fig. 5 shows UFP dose for the periods when concentrations were the highest but also the lowest (i.e. termed as corrected in figure caption). These corresponded to early mornings (before 8 a.m.) and later evening hours (around 8 p.m.). The estimated doses showed that if exercising had been conducted 1–2 h earlier in the morning or slightly later in the evening the respective doses would have been 25 (S2) to 120% lower. Obviously, the smallest percentage differences (between the “received” and “lowest” scenario dose) were observed at S2, where the most of the subjects exercised at around 19:00 when the respective UFP levels reached almost the daily minima.

Based on the last available statistical data, only 36% of adult Portuguese population practice sport activity (EC, 2014), mainly (67% of them) aiming to improve the general health (DGS, 2017). In terms of the environment, it is noteworthy that in Portugal (as in EU in general) sport activities are mainly conducted outdoors (44%), whereas only 31% use indoor facilities (14% in homes, 17% in health clubs or gyms) (DGS, 2017). Estimated doses for females were lower than of males (considering the same age category). This was mainly due to the different activities patterns between both genders. In Portugal, the frequency of conducting sport activities is much lower than the recommended by WHO (INE, 2016; WHO, 2018). On average (and across all age categories), the overall majority (44%) practices physical exercise 1–2 times per week (INE, 2016). Nevertheless, if considering gender and age specific patterns, sport activities are more frequently and during longer duration conducted by males (18 to <35 yrs old) than females (>5 h per week vs. < 2 h; INE, 2016) thus resulting in higher UFP dose. In that regard, it needs to be also noted that estimated UFP doses are influenced by subject-dependent parameters (such as age, breathing rate, body weight, levels of physical conditions, and type of inhalation), which could not be directly assessed for the respective population but retrieved from USEPA (USEPA, 2011). Gender and age-specific values established specifically for Portuguese population would allow deeper simulations including more complex probabilistic exposures (population-related approaches). Furthermore, in order to correctly assess the health risks from air pollution while exercising, actual personal exposure measurements should be measured as well as individuality of each subjects should be

accounted for. Furthermore, the size distribution of the measured fraction would allow to better assess the particle deposition within the human respiratory system (Hussein et al., 2013; Koivisto et al., 2014, 2018) and would provide deeper analysis of the obtained results.

Finally, about 28% of the Portuguese population >15 yrs (correspond to approximately 2.5 million of inhabitants) use walking as a mean of transportation to work or school (INE, 2016). Evaluating the frequency of walking, every-day walking (i.e. 7 days per week) is the most common one (25–35% vs. 6.7–8.6% for 3–4 times per week). Furthermore, majority of people typically walk less than 30 min (61%), whereas only 14.5% walk for more than 1 h. Thus, for comparison, UFP inhalation dosage was calculated for a general population (Fig. 4c) considering 30 min and 1 h of daily exposure time. Once again, the highest dose dosage was observed at S1, which was the most frequented site by the general population (2–12 times in comparisons with other sites). Thus, considering the adverse health impacts of UFP, strategies to minimize the respective exposures while commuting (on foot) should be developed in order to protect the public health.

5. Conclusions

In order to fulfil the gap regarding the existent knowledge, this study aimed to evaluate UFP levels while conducting outdoor physical activities. Across the four sites (S1–S4) regularly used for exercising, UFP number concentration in air highly varied (medians range of $5.1\text{--}20.0 \times 10^3 \text{ \# cm}^{-3}$). Overall, the highest concentrations of UFP were observed at sites which were next to trafficked streets, whereas site S3 and S4 exhibited 2–4 times lower UFP levels. On international level, obtained UFP concentrations were comparable with other countries.

UFP concentrations were significantly correlated ($p < 0.01$) with meteorological parameters. Positive associations between UFP, temperature and solar radiations were most likely due to the atmospheric photochemistry (ozone-initiated processes and photochemistry). In agreement, temporal profiles of UFP demonstrated elevated midday number concentrations, and were probably associated with nucleation processes mediated by photochemistry (Reche et al., 2014; Wang et al., 2010). As increased ambient temperature may lead to elevated air pollution (either aerosols or ozone), in terms of outdoor sport activity exercising early in the morning (especially in warmer seasons) might oppose some of the negative health risks of polluted air.

Considering the WHO recommendations (WHO, 2018), the means of estimated inhalation doses were $1.73 \times 10^8\text{--}3.81 \times 10^8 \text{ \# kg}^{-1}$ for scenarios of moderate activities, and $1.93 \times 10^8\text{--}5.95 \times 10^8 \text{ \# kg}^{-1}$ for the highly-intense ones, with the lowest doses estimated for city park ($8.67 \times 10^7\text{--}1.69 \times 10^8 \text{ \# kg}^{-1}$). The scenarios of highly-intense activities (such as running) led to 2.4 times higher UFP dose for which children and youths (5–17 yrs old) experienced 203–267% higher doses. Considering the national activity patterns, and the age- and gender- differences, the estimated UFP doses of males were 1.1–2.8 times higher than for females, namely for younger populations (18 to <21 and 21 to <30 yrs old). UFP inhalation dose due to scenarios of walking (while commuting to work and or schools) were 1.6–7.5 times lower than when conducting the sport activities. Whereas in clean environments the benefits of physical activity undoubtedly outweigh the risks of air pollution (even with high intense activities), when exercising in polluted atmosphere the harms may exceeded the positive aspects of exercising (Tainio et al., 2016). While national and public programs need to promote healthy and physically active life style of all citizens, strategies to minimize the negative impact of air pollution while exercising should be developed and implemented to best protect

sport practitioners' health.

Ethical statement

The authors declare that there is no conflict of interest regarding the publication of this work.

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Appendix A. Supplementary data

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