ULTRAFINE PARTICLES IN AMBIENT AIR OF AN URBAN AREA: DOSE IMPLICATIONS FOR ELDERLY

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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 study aimed to (1) determine the levels of outdoor pollutants in an urban area with emphasis on UFP concentrations and (2) estimate the respective dose rates of exposure for elderly populations. UFP were continuously measured over 3 weeks at 3 sites in north Portugal: 2 urban (U1 and U2) and 1 rural used as reference (R1). Meteorological parameters and outdoor pollutants including particulate matter (PM_{10}), ozone (O_3), nitric oxide (NO), and nitrogen dioxide (NO_2) were also measured. The dose rates of inhalation exposure to UFP were estimated for three different elderly age categories: 64–70, 71–80, and >81 years. Over the sampling period levels of PM_{10}, O_3 and NO_2 were in compliance with 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 levels were 20–70% lower (mean of 1×10^4 particles/cm^3). Vehicular traffic and local emissions were the predominant identified sources of UFP at urban sites. In addition, results of correlation analysis showed that UFP were meteorologically dependent. Exposure dose rates were 1.2- to 1.4-fold higher at urban than reference sites with the highest levels noted for adults at 71–80 yr, attributed mainly to higher inhalation rates.

Particulate matter (PM) is recognized as one of the most important air pollutants. Epidemiological studies demonstrated an association between increase morbidity and mortality rates due to respiratory and cardiovascular diseases and elevated levels of ambient PM (Brunekreef et al., 2009; Krewski et al., 2003, 2009; Krewski and Rainham, 2007; Samet and Krewski, 2007; Chiu and Yang, 2009; Turner et al., 2011). As evidence has been overwhelming International Agency for Research on Cancer (IARC) in October 2013 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 found that atmospheric particles of different sizes may be responsible for different levels of adverse effects (Su et al., 2006). The smallest fraction of PM is ultrafine particles (UFP) with particle size less than 100 nm (Wang et al., 2011). Unlike larger particles, UFP produce adverse health effects even at low mass levels because of (1) high number concentrations, (2) high specific surface area, and (3) ability to penetrate into pulmonary interstitial spaces (Bakand et al., 2012; Oberdörster et al., 2005; Sioutas et al., 2005). Studies showed that exposures to UFP are associated with impaired lung function and altered 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 underlying UFP-induced adverse health effects are yet to be fully understood. Although epidemiological studies on UFP are needed, exposure assessment issues for UFP are complex, such as high spatial variability and high seasonal variability in UFP number concentration and composition, and these need to be considered prior to undertaking investigation of UFP-mediated adverse health effects (Sioutas et al., 2005).

UFP originate from both natural and anthropogenic sources being emitted as primary origin as well secondarily generated 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, investigators demonstrated that UFP number concentrations and size distribution are also governed by meteorology, thus creating varying patterns (Pirjola et al., 2006; Hussein et al., 2006). In order to fully comprehend these complexities, further studies are needed.

The number of elderly people (i.e., >65 yr) is increasing globally. Between 1996 and 2008 the elderly population rose from 380 to 500 million, that is from 7 to 16% of the total population (Bentayeb et al., 2012). According to the 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, is needed, particularly for the elderly. Compared to the remaining population, the elderly are potentially highly susceptible to effects of outdoor air pollution.

Thus, the present study aimed to determine the levels of outdoor pollution at an urban area with emphasis on UFP. The specific objectives of this investigation were to (1) measure UFP number concentrations at two urban and one rural site (used as reference) and (2) estimate the respective dose rates of inhalation exposure to UFP for elderly populations compared to active adults. The outdoor pollutants such as particles with aerodynamic diameter below 10 μm (PM_{10}) ozone (O_3), nitric and nitrogen oxides (NO and NO_2), and meteorological parameters including temperature (T), relative humidity (RH), wind speed (WS), precipitation (P), and solar radiation (SR), were determined in order to characterize outdoor pollution and weather conditions, and the influence of these conditions on UFP levels.

MATERIALS AND METHODS

Study Area Description

Oporto is the second largest city of Portugal, located in the north of Portugal. The climate is characterized by annual average temperature of approximately 15°C and the difference between the highest and lowest monthly averages being less than 10°C. Annual air humidity is between 75 and 80%, and total annual mean precipitation varies between 1000 mm and 1200 mm, with about 40% in the winter season. Prevailing winds are from west and northwest (Pereira et al., 2007). The important air pollution sources in the respective area are vehicular 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 over 3 weeks in May–June 2013 at 3 different sites

in Portugal. The three sites were selected in order to represent different environments. Sites U1 and U2 were characterized as urban and were situated in the Paranhos district of Oporto city. Previously, Slezakova et al. (2011, 2013) demonstrated that vehicular traffic emissions are the main pollution source in this area. 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, etc.). The third site, R1, was situated in Ermesinde district, also in the north of Portugal. This site was considered as a rural background 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., Shoreview, MN). Intake flow was 0.7 L/min and UFP logging interval was 60 s. Samplers were positioned in open areas avoiding any obstacles and barriers (trees, bushes walls, and fences) that might interfere with data collection. The equipment was mounted on supports (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 over two consecutive weekdays. The number of road vehicles was manually counted for 10 min of each hour between 5:00 and 24:00 h.

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

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 Sites (U1, U2) and the Rural (R1) Site

	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)	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} ($\mu g/m^3$)	25 (14–30)[b]	17 (10-25)[b]	5 (4–6)[b]
O_3 ($\mu g/m^3$)	60 (4–111)	59 (12–100)	53 (32–86)
NO ($\mu g/m^3$)	34 (2–224)	15 (2–129)	1.6 (1.3–2.1)
NO_2 ($\mu g/m^3$)	50 (10–134)	29 (8-83)	1.6 (0.5–5.4)

Note. The means are averaged over 24 h, whereas ranges (in parentheses) represent ranges of 5 min for meteorological parameters and 1-h mean for air pollutants.

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

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

Dose Rate Exposure Analysis

UFP dose rates from inhalation exposure of the elderly were calculated using Eq. (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/day); BR is the age-specific breathing rate (L/min); BW is age-specific body weight (kg); C is the concentration of UFP (number of particles/L); OF is the occupancy factor (i.e., percent likely to be in the public garden at a given interval of time). UFP dose rates were estimated for elderly, that is, adults >65 yr old. The information on age-specific factors was retrieved from the U.S. Environmental Protection Agency (EPA) *Exposure Factors Handbook* (U.S. EPA, 2011) using BW of 72 kg. BR rates corresponding to sedentary activities (which were the mostly observed) were used as the following (U.S. EPA, 2011): 4.9 L/min for

seniors 65–70 yr, 5 L/min for seniors 71–80 yr, and 4.9 L/min for seniors >81 yr. 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 yr) considering the same exposure time (i.e., 2.5 h/d) as for elderly. Age-specific parameters of 4.6 L/min for BR and BW of 76 kg were used for this group (U.S. EPA, 2011).

Statistical Analysis

For data treatment, Student's t-test was applied to determine the statistical significance ($p < .05$, two-tailed) of the differences between the determined means. Spearman's rank correlation coefficient ($p < .05$) 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 and 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 $1.7 \times 10^4 \pm 0.5 \times 10^4$) at U1 and from 2.4×10^3 and 3×10^4 at U2 (mean of $1.2 \times 10^4 \pm 0.6 \times 10^4$). At the rural site, lower levels of UFP were observed with concentrations ranging between 1.5×10^3 and 3.4×10^4 (mean of $1 \times 10^4 \pm 0.7 \times 10^4$). Statistical analysis of these results indicated that (i) UFP concentrations were significantly higher at the urban than rural site, and (ii) differences noted between UFP means at sites U1 and U2 were significant.

The daily profiles of UFP number concentrations at the three sites are shown in Figures 2A–2C, which also demonstrated profiles of traffic density. The average traffic density of the roads around U1 was 16 vehicles/min and traffic peak hours were

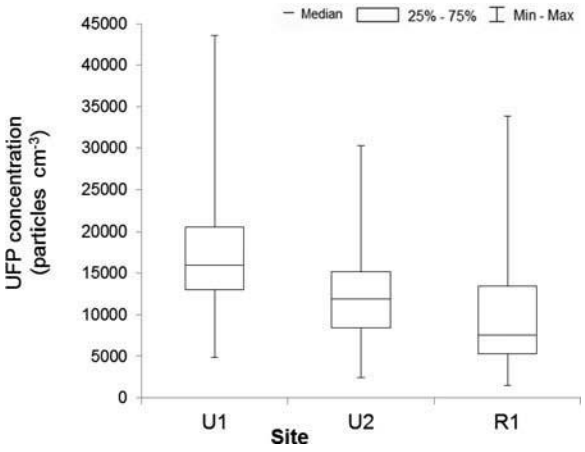

FIGURE 1. UFP number concentrations at two urban sites (U1, U2) and rural (R1) site: minimum and maximum values, median, 25th and 75th percentiles.

detected at 08:30 (24 vehicles/min) and 17:30 hr (25 vehicles/min). All roads around U1 were characterized by type of vehicles, which were mostly cars (95%). Traffic density around site U2 was comparable with U1 (daily average of 13 vehicles/min; traffic peak hours at 08:30 and 18:30 with 21 and 19 vehicles/min, respectively); however, the type of vehicle traffic was different. U2 was situated near a road with a proportion of heavy duty vehicles of 15%, typically buses. As expected, traffic density at R1 was lower than at the other sites. Low traffic density (<1 vehicle/min) 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, daily profiles of UFP at both urban sites also differed to 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 analyzed together with meteorological parameters. Table 2 shows Spearman´s correlation coefficients between UFP number concentrations at sites U1, U2, and R1 and meteorological parameters such as temperature, relative humidity, wind speed, and solar radiation. Inverse correlations between number

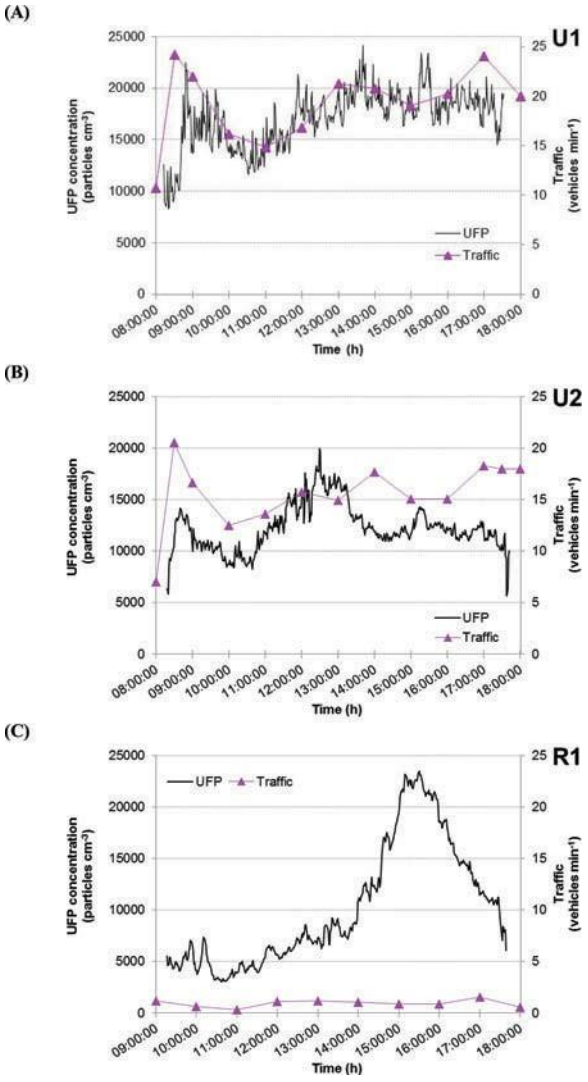

FIGURE 2. UFP number concentrations profiles: (A) urban site U1; (B) urban site U2; and (C) rural site R1. The traffic density profile (between 08:00 and 18:00) at each site is also shown.

of UFP, RH, and WS were noted. Temperature and SR 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 elderly at the three studied sites are presented in Table 3. 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 of age 71–81 yr.

TABLE 2. Spearman Correlation Coefficients Between UFP Number Concentration and Meteorological Parameters at the Two Urban Sites (U1, U2) and the Rural (R1) Site

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

Note. All values are statistically significant for $p < .05$.
[a]Cannot be computed because precipitation was constant (mm) during all the period of the sampling campaign.

DISCUSSION

As humans are adversely affected by exposure to air pollutants in ambient air, the European Union (EU) established health-based standards for a number of pollutants in air under Directive 2008/50/EC (European Union, 2008). These standards are applied over differing periods of time because the observed health impacts associated with various pollutants may occur over different exposure times. Currently 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, PM_{10}, NO_2, and O_3. For O_3, EU set the legislation standard as a maximal daily 8-h mean with limit value of 120 μg/m^3. For NO_2 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 (14–30 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 NO_2 were lower than EU limits, as were the concentrations of O_3. Therefore, over the sampling campaign, levels of the air pollutants were in compliance with EU legislation.

The concentrations of UFP were significantly higher at two urban sites than at rural

TABLE 3. Estimated Dose Rates of UFP (particles/kg/day) for Four Different Age Categories at the Two Urban Sites (U1, U2) and the Rural (R1) Site

		Dose rate (particles/kg)		
		U1	U2	R1
Elderly	65-70 yr	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 yr	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 yr	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 yr	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)

sites. Specifically, UFP levels were, respectively, 70 and 20% higher at U1 and U2 than at R1. Number concentrations of UFP in ambient air varied 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 locations to tunnels with levels ranging from 3×10^3 to 2×10^5 particles/cm^3. Specifically for urban sites, estimated means between 7.2×10^3 and 10.7×10^3 particles/cm^3 (based on 24 studies) were reported. In addition, 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 in a range similar to that noted by Morawska et al (2011). For the European urban sites the latter study reported mean values between 1.2×10^4 (Helsinki, Finland) and 1.9×10^4 particles/cm^3 (Birmingham, UK). It is possible 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) may be attributable to 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 close proximity of the sampling site to the traffic road at U1 (about 8 m) might account for some of these differences (Seigneur, 2009; Sioutas et al. 2005; Solomon 2012). For rural sites, information is available only in the study of Morawska et al. (2008), which estimated a mean of 0.48×10^4 particles/cm^3 (based on 8 studies), which is approximately twofold lower than in the present study. Atmospheric formations of UFP and natural emissions from vegetation such as plantations and forests that were located in the direct vicinity of the site R1 might elevate levels (Morawska et al., 2008). In addition, 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 plowing 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 a rise in UFP and may account for some of these increased UFP levels at R1.

Comparing the two urban sites, the daily profiles of UFP also differed. At site U1 the daily profile of UFP number concentrations was similar to other urban areas (Solomon et al., 2008; Wang et al., 2011). The peaks of UFP number concentrations and traffic density were observed in the same period (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 (data not shown). Therefore, it is possible that UFP number concentrations originated mainly from traffic emissions at this site, owing to the high levels in the morning and 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 noted in in the morning during the rush hour and was associated with motor vehicle emissions. However, the second peak was observed at mid-day to early afternoon (between 12:00 and 13:00). This increase was associated with emissions of the local soup kitchen that was situated closely to U2. Therefore, overall levels of UFP at site U2 resulted from contributions of both these sources.

The results of Spearman correlations showed that coefficients between concentrations of UFP and meteorological parameters, namely, T, RH, WS, and SR, were significant for all variables. The WS displayed a negative correlation with UFP number concentrations due to the greater horizontal dispersion of pollutants at higher WS (Shi et al., 2007). An inverse correlation between UFP and RH may be attributed to the fact that particles are removed from atmosphere by dissolution in water droplets (Agudelo-Castañeda et al., 2013) or by coagulation of droplets on the particles, and thus, easily removed by below-cloud or in-cloud processes (Wiegand et al., 2011). The positive correlation between UFP number concentration, T, and SR might be due to photochemical activity, leading to an increase in concentration of UFP (Park et al., 2008). Specifically, elevation in T produces a rise of tropospheric O_3 (Elminir 2005). The presence of sunlight then increases photolysis of tropospheric O_3 and creates OH radicals that 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 correlation analysis between UFP and meteorological parameters are 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.

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

The dose rates of UFP estimated in this investigation were due to outdoor exposure only. However, individuals 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 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 UFP suggests that considerable efforts are needed in order to understand the linkage between UFP exposures and various types of health outcomes.

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REFERENCES

Agudelo-Castañeda, D. M., Teixeira, E. C., Rolim, S. B. A., Pereira, F. N., and Wiegand, F. 2013. Measurement of particle number and related pollutant concentrations in an urban area in South Brazil. *Atmos. Environ*. 59: 30–38.

Bakand, S., Hayes, A., Dechsakulthorn, F. 2012. Nanoparticles: A review of particle toxicology following inhalation exposure. *Inhal. Toxicol*. 24: 125–135.

Bentayeb, M., Simoni, M., Baiz, N., Norback, D., Baldacci, S., Maio, S., Viegi, G., and Annesi-Maesano, I. 2012. Adverse respi- ratory effects of outdoor air pollution in the elderly. *Int. J. Tuberc. Lung D*. 16: 1149–1161.

Brunekreef, B., Beelen, R., Hoek, G., Schouten, L., Bausch-Goldbohm, S., Fischer, P., Armstrong, B., Hughes, E., Jerrett, M., and van den Brandt, P. 2009. Effects of long-term exposure to traffic-related air pollution on respiratory and cardiovascular mortality in the Netherlands: the NLCS-AIR study. *Res. Rep. Health Effects Inst*. 139: 5–71.

Buonanno, G., Stabile, L., and Morawska, L. 2014. Personal exposure to ultrafine particles: The influence of time–activity patterns. *Sci. Total Environ*. 468–469:903–907.

Castro, D., Slezakova, K., Delerue-Matos, C., Alvim-Ferraz, M. C., Morais, S., and Pereira, M. C. 2011. Polycyclic aromatic hydrocarbons in gas and particulate phases of indoor environments influenced by tobacco smoke: Levels, phase distributions, and health risks. *Atmos. Environ*. 45: 1799–1808.

Chiu, H.-F., and Yang, C.-Y. 2009. Air pollution and emergency room visits for arrhythmias: Are there potentially sensitive groups? *J. Toxicol. Environ. Health A* 72: 817–823.

Elminir, H. K. 2005. Dependence of urban air pollutants on meteorology. *Sci. Total Environ*. 350: 225–237.

European Union. 2008. Directive 2008/50/EC of the European Parliament and of the Council on ambient air quality and cleaner air for Europe. *Off. J. Eur. Union* L152: 1–44. Eurostat. 2013. Population structure and ageing. http://epp.eurostat.ec.europa.eu/statistics_explained/index.php/Population_structure_and_ageing (accessed October 2013).

Ferreira, A. J., Cemlyn-Jones, J., and Robalo Cordeiro, C. 2013. Nanoparticles, nanotechnology and pulmonary nanotoxicology. *Rev. Port. Pnemol*. 19: 28–37.

Hussein, T., Karppinen, A., Kukkonen, J., Harkonen, J., Aalto, P. P., Hameri, K., Kerminen, V.-M., and Kulmal, M., 2006. Meteorological dependence of size fractionated number concentrations of urban aerosol particles. *Atmos. Environ*. 40: 1427–1440.

International Agency for Research on Cancer. 2013. The carcinogenicity of outdoor air pollution. *Lancet Oncol*. doi:10.1016/S1470-2045(13)70487-.

Kalaiarasan, M., Balasubramanian, R., Cheong, K. W. D., and Tham, K. W. 2009. Traffic-generated airborne particles in naturally ventilated multi-storey residential buildings of Singapore: Vertical distribution and potential health risks. *Build. Environ*. 44: 1493–1500.

Kanawade, V. P., Benson, D. R., and Lee, S.-H. 2012. Statistical analysis of 4-year observations of aerosol sizes in a semi-rural continental environment. *Atmos. Environ*. 59: 30–38.

Krewski, D., Burnett, R. T., Goldberg, M. S., Hoover, B. K., Siemiatycki, J., Jerrett, M., Abrahamowicz, M., and White, W. H. 2003. Overview of the reanalysis of the Harvard Six Cities Study and American Cancer Society study of particulate air pollution and mortality. *J. Toxicol. Environ. Health A* 66: 1507–1551.

Krewski, D., Jerrett, M., Burnett, R. T., Ma, R., Hughes, E., Shi, Y., Turner, M. C., Pope 3rd., C. A., Thurston, G., Calle, E. E., Thun, M. J., Beckerman, B., DeLuca, P., Finkelstein, N., Ito, K., Moore, D. K., Newbold, K. B., Ramsay, T.,

Ross, Z., Shin, H., and Tempalski, B. 2009. Extended follow-up and spatial analysis of the American Cancer Society study linking particulate air pollution and mortality. *Res. Rep. Health Effects Inst*. 140: 5–114.

Krewski, D., and Rainham, D. 2007. Ambient air pollution and population health: Overview. *J. Toxicol. Environ. Health A* 70: 275–283.

Kumar, P., Robins, A., Vardoulakis, S., and Britter, R. 2010. A review of the characteristics of nanoparticles in the urban atmosphere and the prospects for developing regulatory controls. *Atmos. Environ*. 44: 5035–5052.

Kumar, P., Robins, A., Vardoulakis, S., and Quincey, P. 2011. Technical challenges in tackling regulatory concerns for urban atmospheric nanoparticles. *Particuology* 9: 566–571.

Maynard, R., Krewski, D., Burnett, R., Samet, J., Brook, J., Granville, G., and Craig, L. 2003. Health and air quality: Directions for policy-relevant research. *J. Toxicol. Environ. Health A* 66: 1891–1904.

Morawska, L., Ristovski, Z., Jayaratne, E. R., Keogh, D. U., and Ling, X. 2008. Ambient nano and ultrafine particles from motor vehicle emissions: Characteristics, ambient processing and implications on human exposure. *Atmos. Environ*. 42: 8113–8138.

Oberdörster, G. 2001. Pulmonary effects of inhaled ultrafine particles. *Int. Arch. Occup. Environ. Health* 74: 1–8.

Oberdörster, G., Oberdörster, E., and Oberdörster, J. 2005. Nanotoxicology: An emerging discipline evolving from studies of ultrafine particles. *Environ. Health Perspect*. 113: 823–839.

Park, K., Park, J. Y., Kwak, J.-H., Cho, G. N., and Kim, J.-S. 2008. Seasonal and diurnal variations of ultrafine particle concentration in urban Gwangju, Korea: Observation of ultrafine particle events. *Atmos. Environ*. 42: 788–799.

Pereira, M. C., Santos, R. C., and Alvim-Ferraz, M. C. M. 2007. Air quality improvements using European environment policies: A case study of SO_2 in a coastal region in Portugal. *J. Toxicol. Environ. Health A* 70: 1–5.

Pirjola, L., Paasonen, P., Pfeiffer, D., Hussein, T., Hameri, K., Koskentalo, T., Virtanen, A., Ronkko, T., Keskinen, J., Pakkanen, T. A., and Hillamo, R. E., 2006. Dispersion of particles and trace gases nearby a city highway: Mobile laboratory measurements in Finland. *Atmos. Environ*. 40: 867–879.

Samet, J., and Krewski, D. 2007. Health effects associated with exposure to ambient air pollution. *J. Toxicol. Environ. Health A* 70: 227–242.

Seigneur, C. 2009. Current understanding of ultrafine participate matter emitted from mobile sources. *J. Air Waste Manage. Assoc.* 59: 3–17.

Shi, Z., He, K., Yu, Z., Yao X., Yang, F., Ma, Y., Ma, R., Jia, Y., and Zhang, J. 2007. Diurnal variation of number concentration and size distribution of ultrafine particles in the urban atmosphere of Beijing in winter. *J. Environ. Sci*. 19: 933–938.

Sioutas, C., Delfino, R., and Singh, M. 2005. Exposure assessment for atmospheric ultrafine particles (UFPs) and implications in epidemiologic research. *Environ. Health Perspect*. 113: 947–955.

Slezakova, K., Castro, D., Begonha, A., Delerue-Matos, C., Alvim-Ferraz, M. C., Morais, S., and Pereira, M. C. 2011. Air pollution from traffic emissions in Oporto, Portugal: Health and environmental implications. *Microchem. J*. 99: 51–59.

Slezakova, K., Pires, J. C. M., Castro, D., Alvim-Ferraz, M. C. M., Delerue-Matos, C., Morais, S., and Pereira, M. C. 2013. PAH air pollution at a Portuguese urban area: Carcinogenic risks and sources identification. *Environ. Sci. Pollut. Res. Int*. 20: 3932–3945.

Solomon, P. A. 2012. An overview of ultrafine particles in ambient air. *EM* May: 18–23.

Solomon, P. A., Hopke, P. K., Froines, J., and Scheffe, R. 2008. Key scientific findings and policy- and health-relevant insights from the U.S. Environmental Protection Agency's Particulate Matter Supersites Program and related studies: an integration and synthesis of results. *J. Air Waste Manage. Assoc.* 58(13 suppl.): S3–S92.

Stanek, L. W., Sacks, J. D., Dutton, S. J., and Dubois, J. J. B. 2011. Attributing health effects to apportioned components and sources of particulate matter: An evaluation of collective results. *Atmos. Environ*. 45: 5655–5663.

Su, Y., Sipin, M. F., Spencer, M. T., Qin, X., Moffet, R. C., Shields, L. G., Prather, K. A., Venkatachari, P., Jeong, C.-H., Kim, E., Hopke, P. K., Gelein, R. M., Utell, M. J., Oberdörster, G., Berntsen, J., Devlin, R. B., and Lung, C. C. 2006. Real-time characterization of the composition of individual particles emitted from ultrafine particle concentrators. *Aerosol Sci. Technol*. 40: 437–455.

Turner, M. C., Krewski, D., Pope, C. A. III, Chen, Y., Gapstur, S. M., and Thun, M. J. 2011. Long-term ambient fine particulate matter air pollution and lung cancer in a large cohort of never-smokers. *Am. J. Respir. Crit. Care Med*. 184: 1374–1381.

United Nations, Department of Economics and Social Affairs, Population Division 2001. *World population ageing 1950–2050*. ST/ESA/SER.A/207. New York, NY: United Nations.

U.S. Environmental Protection Agency. 2011. *Exposure factors handbook:* 2011 *Edition*. EPA/600/R–09/052F. Washington, DC: U.S. EPA Office for Research and Development.

Wang, F., Costabileb, F., Li, H., Fang, D., and Alligrini, I. 2010. Measurements of ultrafine particle size distribution near Rome. *Atmos. Res*. 98: 69–77.

Wang, Y., Hopke, P. K., Chalupa, D. C., and Utell, M. J. 2011. Long-term study of urban ultrafine particles and other pollutants. *Atmos. Environ*. 45: 7672–7680.

Wiegand, F., Pereira, F. N., and Teixeira, E. C. 2011. Study on wet scavenging of atmospheric pollutants in south Brazil. *Atmos. Environ*. 45: 4770–4776.