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Measurement of airborne ultrafine particles in work and life environments: study design and preliminary trends in an Italian university site.

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Abstract. Airborne ultrafine particles (UFPs) potentially have adverse health effects for the exposed population both in life and work environments. Main objective of this study is to propose a strategy to measure multiple parameters that may influence human exposure to UFPs and apply it in a case study at the university site of "La Sapienza" in Rome (Italy). The measurements allow the characterization of UFPs in terms of particle number concentration (PNC), particle size distribution (PSD), lung deposited surface area (LDSA), geometric mean diameter (GMD) and polycyclic aromatic hydrocarbons concentration (PAHs), using data collected in indoor and outdoor conditions by real-time instruments with high time resolution. The study design includes: i) inter-comparison of devices based on different principles of operation to harmonize the outputs in relation to a reference instrument; ii) case study site description and sampling points identification to allow indoor vs outdoor relations at different height; iii) extensive measurement campaigns to provide a dataset useful to data comparison; iv) statistical data series analysis. The preliminary results reported here are related to the values of PNC and calculated average diameter (D_{avg}) for airborne UFPs, obtained in three different sampling points of the research workplaces at the university site: auditorium (about 300 places - ground floor), computer science laboratory (about 50 workstations - first floor) and external roof (top of the building). Major relations between PNC and D_{avg} have been highlighted in daily trends.

Introduction
Airborne ultrafine particles (UFPs) are defined as particles with aerodynamic diameter less than 0.1 μm [1]; UFPs may be classified in natural, e.g. atmospheric or marine aerosol [2], and anthropogenic, typically generated through combustion of biomass (e.g., tobacco smoking, wood burning, incense burning) or fossil fuels (e.g., coal, natural gas, diesel) [3]. In the last years, the increasing interest in nanotechnology research, introduced further sources of particles that may fall under UFPs definition due to their size: deliberately produced engineered nanomaterials and incidental nanoparticles as a result of multiple process in manufacturing and service industries as intermediates, by-products and waste emissions [4]. Many studies have shown that UFPs can contribute to adverse health effects in living and work environments [5-8]. Research questions are still open about UFPs metrics and indicators of interest that might be used to characterize potential emissions, air quality, exposure and health effects [9]. Studies on occupational exposure to nanomaterials highlighted also the need to apply a multi-metric approach to study the potential health effects, to identify the emission sources and better characterize the background [10-11].

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In this context, the main objective of this study is to propose a strategy to measure multiple parameters that may influence human exposure to UFPs; we applied such strategy in a case study at the university site of "La Sapienza" in Rome (Italy) in research workplaces. Preliminary results of in situ measurement campaigns are reported here in daily trends.

Materials and methods

The proposed strategy includes measurements to characterize UFPs in terms of particle number concentration (PNC), particle size distribution (PSD), lung deposited surface area (LDSA), geometric diameter (GMD) and polycyclic aromatic hydrocarbons (PAHs) concentration, using data collected in indoor and outdoor conditions by the following real-time instruments with high time resolution (Table 1):

- Condensation Particle Counter (CPC mod. 3007, TSI Inc., Shoreview, MN, USA) to measure PNC ($\#/cm^3$) of particles from 10 nm to 1 µm, with 1 s time resolution and accuracy ±20% (total flow 0.7 L/min; detection limits 1 to 100,000 $\#/cm^3$).
- Fast Mobile Particle Sizer (FMPS mod. 3091, TSI Inc. Shoreview, MN, USA) to measure SD (dN/dLogDp) and total PNC ($\#/cm^3$), in the size range 5.6–560 nm, with 1 s time resolution; the instrument provides also the atmospheric pressure (bar) and the temperature (°C) at his location.
- Nanoparticle Surface Area Monitor (NSAM mod. 3550, TSI Inc., Shoreview, MN, USA) to measure average and cumulative LDSA ($\mu m^2/cm^3$) of particles from 10 nm to 1 µm corresponding to the tracheobronchial (TB) or alveolar (A) pulmonary fractions (1 s time resolution), based on the model published by the International Commission on Radiological Protection [12].
- Mini Diffusion Size Classifier (DISCmini, mod. TESTO) for the measurement of PNC ($\#/cm^3$), modal diameter (nm) and LDSA ($\mu m^2/cm^3$) with a time resolution of 1 s and accuracy of ±30% (total flow 0.99 L/min; detection limits 1 to 100,000 $\#/cm^3$).
- Electric Low Pressure Impactor (ELPI+, mod. Dekati) to measure SD (dN/dLogDp) and total PNC ($\#/cm^3$), in the size range 6 nm – 10 µm, with 1 s time resolution.
- Photoelectric Aerosol Sensor (PAS2000, EcoChem Analytics, League City, TX USA) to measure PAHs surface-adsorbed on carbon aerosol with aerodynamic diameter from 10 nm to 1.5 µm, with a response time of 10 s in a measuring interval from 0 to 1000 ng/m^3 and a lower detection limit of 3 ng/m^3.

Table 1. Set of instruments included in the general study design and related size range.

	MODEL	*PARAMETER*	*SIZE RANGE*
CPC	3007 TSI	PNC ($\#/cm^3$)	10nm-1µm
FMPS	3091 TSI	PNC ($\#/cm^3$) PSD (dN/dLogDp)	5.6nm-560nm
NSAM	3550 TSI	LDSA* ($\mu m^2/cm^3$)	10nm-1µm
DISCmini	TESTO	PNC ($\#/cm^3$) LDSA ($\mu m^2/cm^3$) GMD** (nm)	10nm-700nm
ELPI+	Dekati	PNC ($\#/cm^3$) PSD (dN/dLogDp)	6nm-10µm
PAS2000	Ecochem	PAH (ng/m^3)	10nm-1.5µm

* running average LDSA for tracheobronchial or alveolar fraction
** GMD in the size range 10nm -300nm

The general study design includes: i) inter-comparison of devices based on different principles of operation to harmonize the outputs in relation to a reference instrument; ii) case study site description and sampling points identification to allow indoor vs outdoor relations at different height (Z/H); iii)

extensive measurement campaigns to provide a dataset useful to data comparison; iv) statistical data series analysis.

The results reported here are related to the extensive winter campaign carried out from November 4th to December 3rd 2017 at the University site (Figure 1). Data of PNC and PSD measured by FMPS for airborne UFPs in the size range 5.6-560 nm, were collected in three different points during a working week: 1. external roof (top of the building Z/H = 1.1) from Nov 13rd to Nov 17th, sampling probes are placed near the center of the terrace; 2. computer science laboratory (about 50 workstations - first floor Z/H = 0.3) from Nov 20th to Nov 24th, sampling probes are placed on the bottom of the room, minimizing influences from the door opening and the heating system; auditorium (about 300 places - ground floor Z/H = 0.1) from Nov 27th to Dec 1st, instruments sampling probes are placed approximately in the center of the room. Night measurement were also performed including non-working activities.

Temperature and pressure values provided by FMPS at the instrument location may serve as control of local weather conditions and climatic influences that may affect the measurements.

Figure 1. University site building and sampling point locations (41°54'05"N 12°30'57"E): 1. Roof top (outdoor), 2. Computer lab first floor (indoor), 3. Auditorium ground floor (indoor). Source: GoogleMaps®, 2019.

D_{avg} calculation were performed according to the following equation [13]:

$$D_{avg}(t) = \frac{\sum_i N_i * d_i}{\sum_i N_i} \qquad (1)$$

Where N_i is the PNC related to the ith diameter d_i, with i ranging from 5.6 to 560 nm (into the 32 different channels of FMPS).

Results

Average 24-h daily trends of total PNC measured by FMPS (size range 5.6-560 nm) for each sampling point are reported in Figure 2. The figure shows that before 7:00am and after 9:00pm the average levels of PNC on the external roof (Z/H = 1.1) are lower than both sampling points inside the building (computer lab and auditorium). On the contrary, during the daylight hours the levels shows a PNC trend highly variable from about 1.30E+04 to 2.00E+04 #/cm³ in all three sampling points.
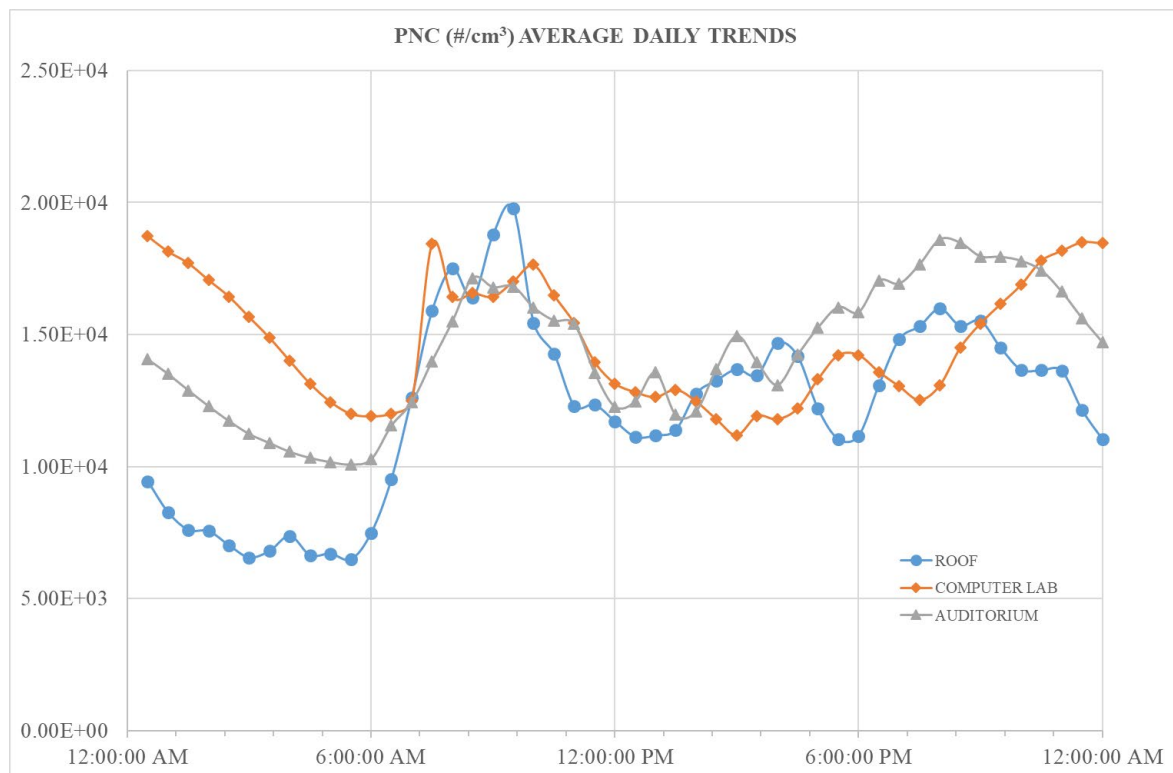
Figure 2. Average 24-h daily trends of total PNC (#/cm^3) in the size range 5.6-560 nm: (blue) roof top of the building outdoor; (orange) computer lab first floor indoor; (grey) auditorium ground floor indoor.

The average 24-h daily trends of D_{avg} calculated according to the (1) for three different sampling points, are reported in Figure 3. D_{avg} of UFPs inside the computer lab is higher than both other two sampling points during the whole average daily trend. Furthermore, starting from the beginning of the day, a negative slope of the curves clearly results for all the sampling points; on the contrary, starting from 5:00pm the slope of three curves reverses and grows up to reach the respective initial value.
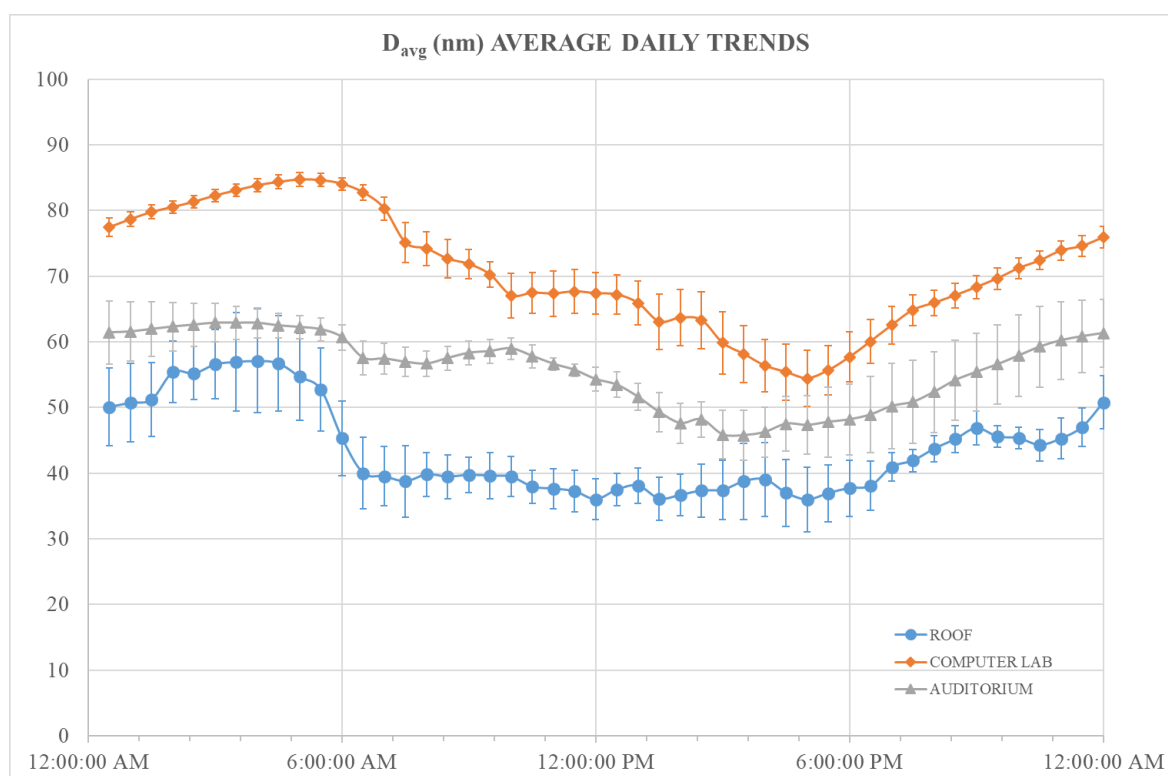
Figure 3. Average 24-h daily trends of D_{avg} (nm) in the size range 5.6-560 nm and related standard errors: (blue) roof; (orange) computer lab; (grey) auditorium.

Discussion and Conclusions

The proposed strategy allows us to characterize UFPs exposure in both work and life environments: the integration of multi-metric data will provide information on different parameters that may influence potential adverse health effects.

In this paper we reported preliminary data related to UFPs PNC measured by FMPS and calculated D_{avg} in the size range from 5.6 to 560 nm. Although the indoor/outdoor measurements were conducted in consecutive periods and not in parallel, some preliminary indications may be highlighted. In general, data trends suggest that multiple factors may influence UFPs PNC levels and their size in the studied workplaces, probably related to both outdoor and indoor contributions.

In particular, at the beginning of the day, in the transition hours (from 5 am to 7 am), we note an increase in outdoor UFPs PNC, probably due to the starting of atrophic activities; the indoor values reflect the same structure with a certain delay time. The same behaviour may be highlighted also in D_{avg} trends, in which the contribution of small particles increases initially on the terrace and after on the other two indoor sampling points.

During the daylight hours (from 7 am to 4 pm) the outside component outnumbers the influences from other internal sources of PNC: in this period the outdoor mean particle size is quite constant, while both the indoor D_{avg} values have negative trends.

In the transition from daylight hours to the night (from 4 pm to 6 pm) a general increase of PNC in indoor sampling points may be mainly recognized; while the average particle size in all the sampling points and periods grows up.

Such data investigation will be useful also to characterize the background for workers involved in activities with potential emission of UFPs from different specific sources, such as engineered nanomaterials research and development processes and others. In this case the identification of reference background levels is crucial to evaluate workers' exposure to the specific process that may generate UFPs and nanomaterials [10].

Relations between indoor and outdoor parameters may be deeply investigated in order to evaluate the outdoor influences to the indoor UFPs levels. Comparison between working and non-working daily trends are useful to highlight the contribution of urban pollutants. Furthermore seasonal variations might be analysed to characterize the climatic influences and the variation of UFP levels produced by heating and cooling systems inside the building.

In conclusion, this study is a part of the INAIL project "VIEPI" (Integrated Evaluation of Indoor Particulate Exposure) in which an extensive data analysis is still ongoing [14]. We feel that the integration of all results that may integrate UFPs findings, including local weather conditions and indoor/outdoor air particles chemical composition, will allow us to obtain a comprehensive characterization of the mechanisms of exposure.

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