Spatial distribution of ultrafine particles at urban scale: the road-to-ambient stage

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Abstract

The spatial variability of ultrafine particles (UFPs) is believed to be an important issue to assess urban air pollution fate and exposure in connection with traffic motorised emissions. In this work, the high-time resolution total number concentration of UFPs was measured at traffic-oriented and urban background locations in a middle-size city in Italy. The major objective was to study valuable connections with local traffic sources, as well as measurement sites' representativeness. On the one hand, it was found that the total concentration at the traffic site can be representative of vehicle exhaust sources in ambient air. On the other hand, it was possible to identify three prevailing contributions for the total UFPs number concentration at urban scale: a very low urban background concentration, a significant contribution due to local traffic sources, and a significant contribution due to secondary transformation processes closely linked to meteorology.

Keywords: urban air pollution, ultrafine particles, traffic emissions, background, representativeness, exposure.

1 Introduction

During the last decades a growing body of research has investigated worldwide the extremely vast subject of urban air quality [1]. Measuring any potential effect of any urban air pollutant requires the understanding of its variation and distribution in both space and time. Traffic-related pollution and its spatial variations are particular concerns; a comprehensive understanding is crucial [2–

WIT Transactions on Ecology and the Environment, Vol 116, © 2008 WIT Press
www.witpress.com, ISSN 1743-3541 (on-line)
doi:10.2495/AIR080531

8]. However, current actions are particularly hampered by a lack of knowledge when characterising the spatial variability within urban areas [9].

For particles, the spatial variability is particularly relevant, and depends on the size fraction. Particles smaller than 100 nm (the so-called ultrafine particles, UFPs) are more variable in space and time than fine particles as they have a higher dependence on particle sources, and a faster removal from the atmosphere [10–12]. Consequently, their spatial variability is believed to be an important issue to assess air pollution fate and exposure.

Internal combustion engines are known to be a major emission source of UFPs [13–17]. However, in spite of extensive laboratory studies on engine emissions, there are few investigations of how particle mobile emissions evolve and affect air quality establishing a link between sources and receptors [18–20]. It has been shown that the size distribution of emitted particles evolves substantially within a few hundred meters of emission [8, 21]. After release into the atmosphere, UFPs are subjected to complex dilution and transformation processes. Neither current models nor those that will be available in the near future are tough to be able to cover all the spatial and temporal scales that are involved from the emission (centimetres, milliseconds) to the urban/regional scale (kilometres, hours) [22].

In this work, high-time resolution UFP total number concentration was measured in a middle-size urban area of Italy (Parma). The major objective was to study valuable connections with local traffic sources, as well as the measurement sites' representativeness [23].

2 Experimental

A Water-Condensation Particle Counter (WCPC,TSI Mod. 3781, Figure 1 and Figure 2) [24–27] was used to measure the concentration of particles in air with a diameter larger than 6 nm (and smaller than 3 μm), $N_{6,}$

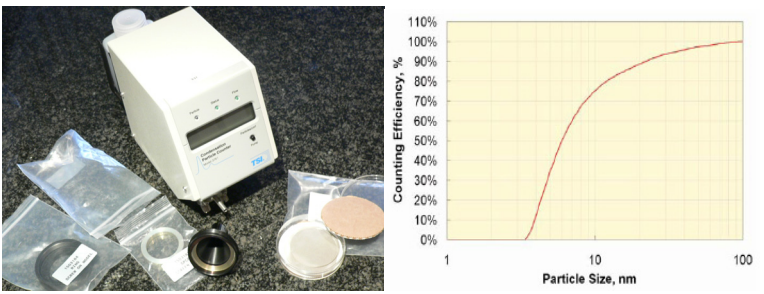

Figure 1: Picture and counting efficiency of the Water-Condensation Particle Counter (WCPC,TSI, Modello 3781) [28].

Data were collected every 2 seconds, and then averaged to generate 1-minute values. Measurements were taken during the winter (January, February and March) and summer (July, August, September) of 2007 (both weekdays and

weekends). Instruments were located at two urban sites, 740 meters away from each other in Parma downtown: via Montebello, and Parco della Cittadella (Figure 3, Figure 4).

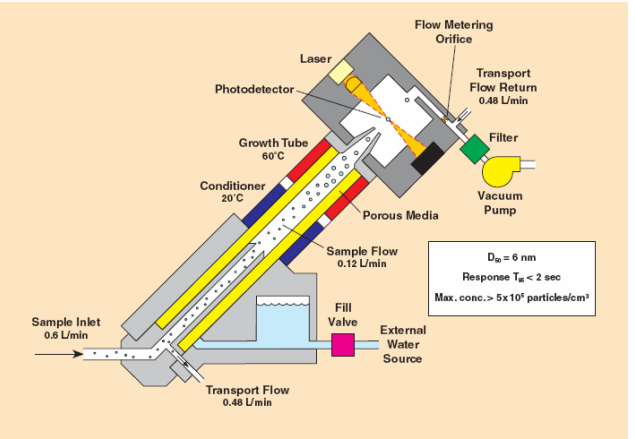

Figure 2: Working principle of the Water-Condensation Particle Counter (WCPC,TSI, Modello 3781) [28].

Figure 3: Picture by Google Earth of the two measurement sites in Parma: Parco della Cittadella (top) and via Montebello (bottom).

Representativeness criteria with respect to emission sources drove the selection of these two sites [29, 30]. Parco della Cittadella was selected as representative of urban background concentrations, the closest traffic emission

source being around 100 meters away. Conversely, the traffic site in Via Montebello would reflect pollution concentrations strongly influenced by urban traffic emissions; measurements were taken less than 1 meter from the road (the road is south of the measurement point, Figure 4). The sampling probe was made by a straight stainless steel tube, with 7 mm internal diameter and length < 15 cm, in order for the sampling losses to be negligible. A small cyclone was used at the traffic site (where the concentrations were expected to be higher) to avoid particles larger than 1 μm to either block or damage the system.

Figure 4: Details by Google Earth of the two measurement sites in Parma: Parco della Cittadella (left) and via Montebello (right).

3 Results and discussion

3.1 From traffic site to urban background

The total number concentrations of UFPs measured (Figures 5–8) were found to be low, but comparable to similar works [31]: urban background locations have usually average concentrations of 10.000 #/cm^3, whereas traffic locations exhibit 30.000-50.000 #/cm^3. Since the total particle number is generally dominated by local emissions, it is particularly affected by primary particles generated by vehicle exhausts. Therefore, as found in previous studies [32], UFP total number concentrations measured at Via Montebello – close to traffic emissions – showed tendencies significantly different from the concentrations measured just 700 meters away in the park; representative of the urban background. Differences related mainly to two factors. The traffic site showed the UFP total number to have both much higher concentrations, and much more rapid fluctuations (Figures 5–8). Much faster and more intensive dilution processes could result in a more rapid reduction of UFP total number, as well as a trigger for other physical processes (such as nucleation) in connection to other parameters, such as solar radiation. Concentrations are comparable only at nighttime, from midnight to 4.00 a.m., when both the traffic flow and the concentration fluctuations at the Montebello station were very low.

Contrary to the urban background site, the total number concentrations at the traffic location showed a daytime trend shaped by two peaks (Figure 10). They can be likely related to traffic rush hours (7.00 a.m. and 18:00), and were also

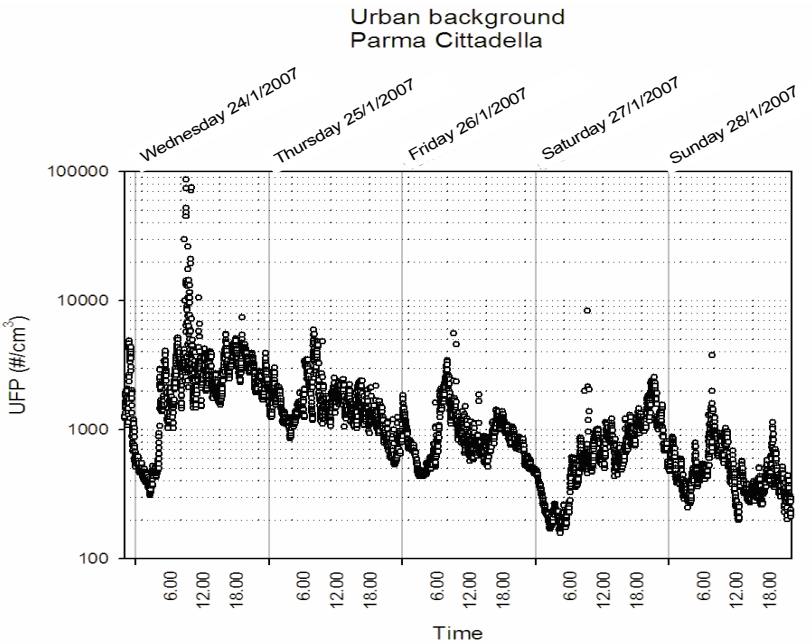

Figure 5: UFP number concentration during the winter campaign at an urban background station in Parma [23].

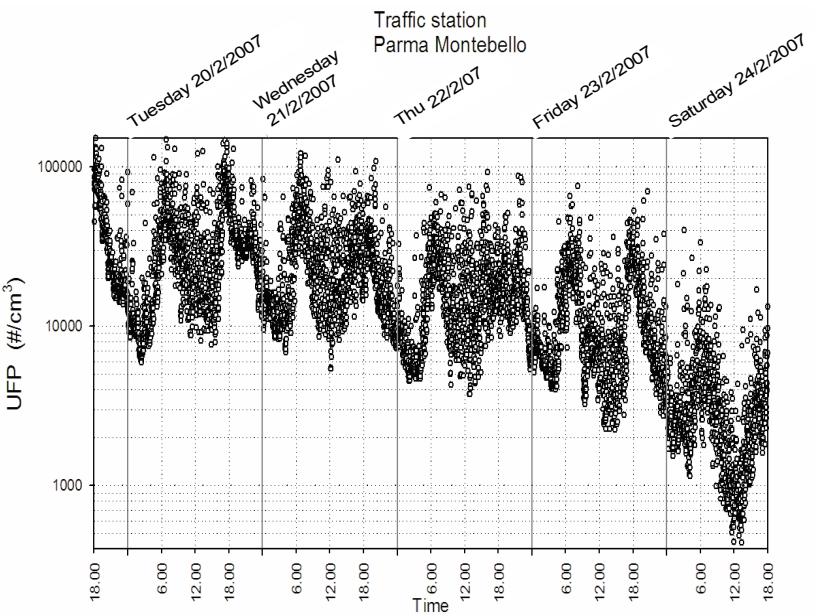

Figure 6: UFP number concentration during the winter campaign at a traffic station in Parma [23].

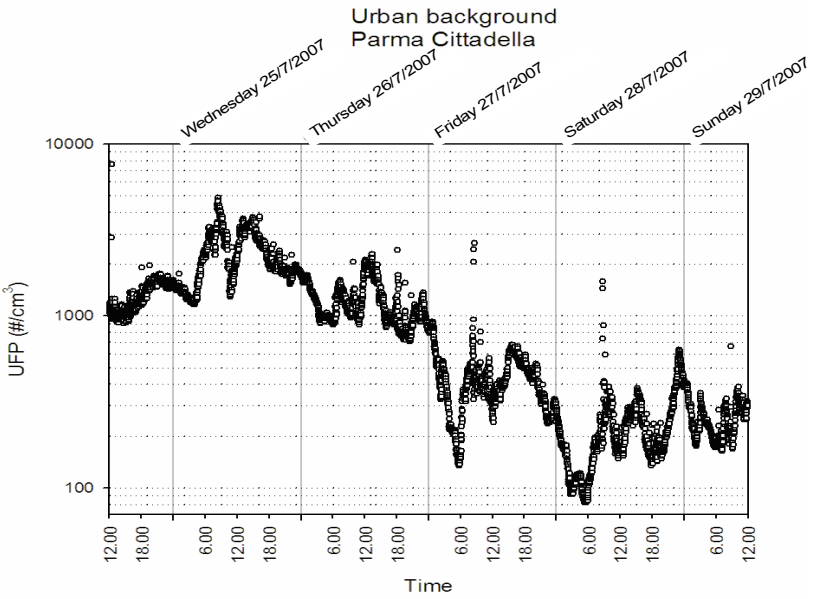

Figure 7: UFP number concentration during the summer campaign at the background station in Parma [23].

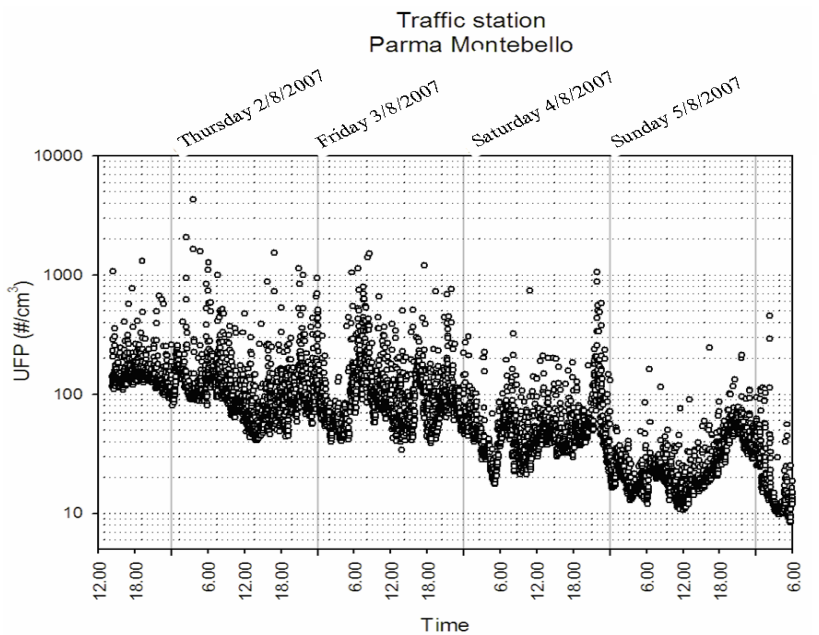

Figure 8: UFP number concentration during the summer campaign at a traffic station in Parma [23].

shown by other gaseous pollutants. This finding clearly indicates traffic emissions to be the prevailing source of the UFP total number [33].

3.2 Coupling with meteorology and gaseous pollutants

Variations due to meteorology can be more easily analysed at the urban background site, where traffic flows influence can, however, also be recognised. UFP total number concentration peaks (relative or absolute peaks) measured in the Cittadella often corresponded to (midday) solar radiation peaks (Figures 5 and 7). This suggests the secondary particle formation to be more significant at the background site [31]. The highest value of UFP total number concentration (not including spikes) at this site was measured on January 19 (Figure 9). This

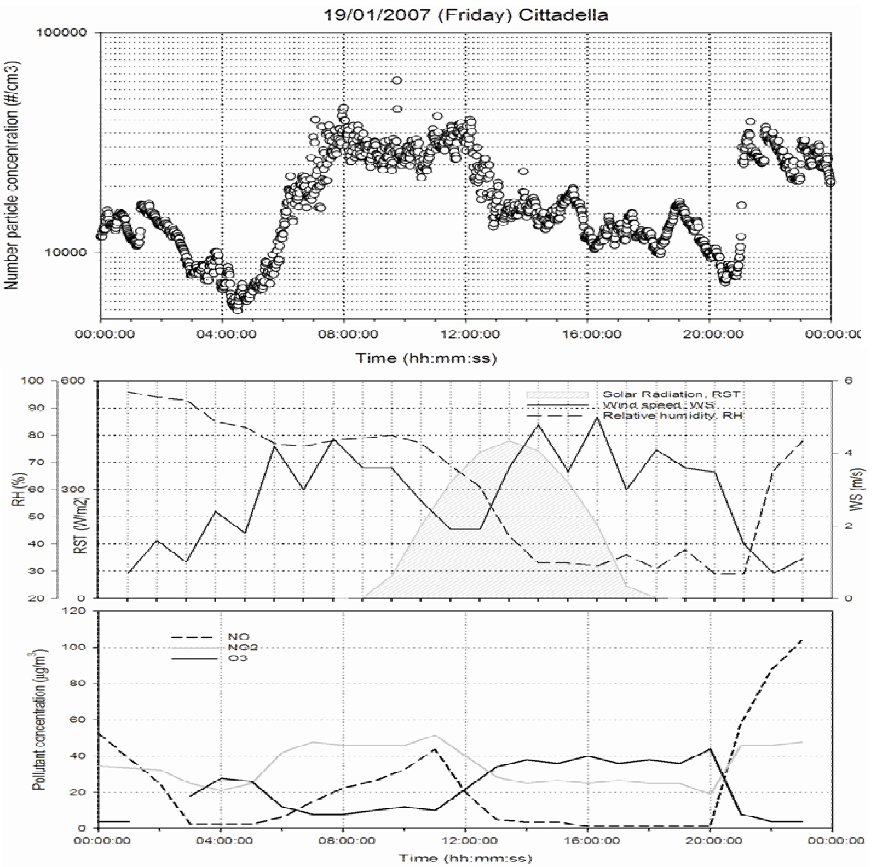

Figure 9: Total number concentration of UFPs measured at the background station (Parco della Cittadella) on Jan 19, 2007. Meteorological parameters and gaseous pollutant concentrations measured at the same site (ARPA Parma) are shown below.

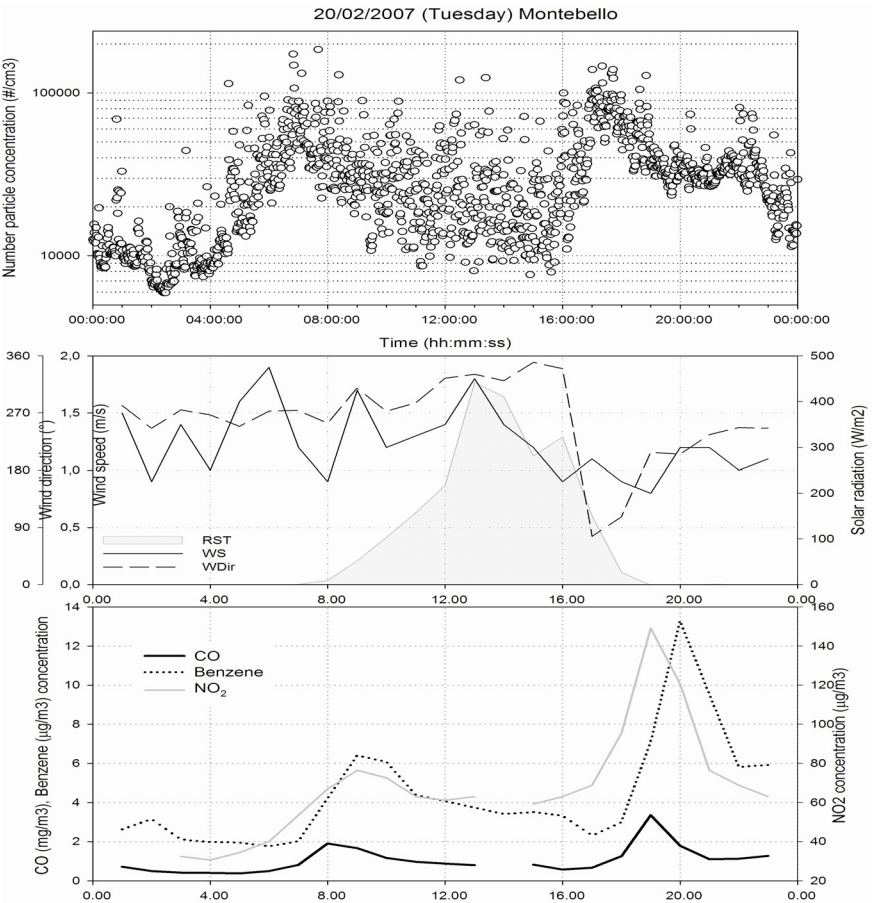

Figure 10: Total number concentration of UFPs measured at the traffic site (via Montebello) on Feb 20, 2007. Meteorological parameters and gaseous pollutant concentrations measured at the same site (ARPA Parma) are shown below.

was a weekday (Friday) with high solar radiation, temperature and wind speed, and low relative humidity.

Though less clear, these noontime peaks can also be seen at the traffic site (Figures 6, 8 and 10).

Beyond this effect, a correlation with wind speed was also found (Figure 9, Figure 10). It is not clear whether increased wind speeds can either induce significant transport phenomena of UFPs emitted/formed by nearby traffic sources [33], or enhance total number concentration by other phenomena (i.e., secondary new particle formation). This effect was less evident at the traffic site: local emitted particles effectively dominated measurements even when the site was upwind to the road nearby. It is not clear if the afternoon peak measured for

both UFPs and gaseous pollutants on February, 20 in Montebello (Figure 10) should be related either to the wind direction change, or (more likely) to the lower wind speed (wind calm, <1m/s), the rush hours, and increased atmospheric stability. Interestingly enough, diurnal trends of both total UFPs number and primary gaseous pollutants (particularly NO and CO) were found to be quite similar (Figures 9 and 10) [34, 35]. Different time resolutions of data collected (1min versus 1h) are likely the major reason for the differences in the fluctuations, and the related time shifts. These findings indicate the total UFP number as a valuable marker of fresh emissions at traffic stations, significantly correlated with combustion-generated pollutants.

Finally, the comparison between the frequency distributions of UFPs measured at the two sites (Figure 11) shows a double FD mode only at the urban background location. This could likely suggest the existence of an (extremely low) urban background level (the lowest mode, 1000#/cm^3, which can be recognised at both the sites). The second mode in the background location could be related to UFPs concentrations (extremely diluted) transported from nearby sources and already aged because of meteorology-induced transformations. The FD at the traffic site looks like an Ln-FD, suggesting mixing and dilution of local traffic source emissions to be the prevailing process over time [29, 36]

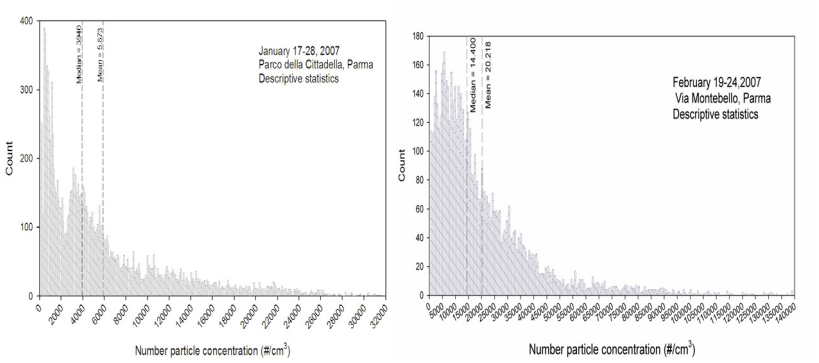

Figure 11: Frequency distribution of the total number concentration of UFPs measured at the background (Parco della Cittadella) and traffic (Via Montebello) stations.

4 Conclusions

The high-time resolution total number concentration of UFPs was analysed at traffic-oriented and urban background locations of a middle-size city in Italy. Total number concentration at the traffic site was found to be a clear indication of vehicle exhaust sources in ambient air. At an urban scale it was possible to identify three prevailing contributions for the total UFPs number concentration. Firstly, a very low urban background concentration (lower than 1000 #/cm^3) was particularly visible in summer. Secondly, a significant contribution due to local

traffic sources, up to 100.000 #/cm^3 (traffic site in winter time). Finally, a significant contribution due to secondary transformation processes closely linked to meteorology, particularly solar radiation. Transport from sources nearby was also found to be significant, indicating significant UFP lifetime in the atmosphere. The conditions near mobile emission sources were found to differ from typical background conditions in that total particle number concentrations were much higher, and dilution processes were much faster and stronger. The stronger variability induced more rapid concentration fluctuations, probably due to faster and more intense dilution processes, which can reduce the number concentration more rapidly, as well as trigger physical processes other than dilution, such as nucleation, coagulation, and condensation.

Acknowledgements

Financial support mainly from the Italian Ministry of Environment, Land and Sea is gratefully acknowledged. As well, the authors would like to thank the ARPA-Parma (Regional Environment Protection Agency), and in particular Eriberto De Munari. Finally, our thanks are extended to our colleagues, F. De Santis, M. Montagnoli, and M. Giusto.

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