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Ultrafine particle apportionment and exposure assessment in respect of linear and point sources

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ABSTRACT

The effects of particulate matter on the environment and public health were widely studied in recent years but agreement amongst these studies on the relative importance of the particle size and its origin with respect to health effects is still lacking. Nevertheless, air quality standards are moving towards greater focus on the smaller particles. In industrialized areas, anthropogenic activities are a major contributor to the particle concentrations. Then, it is important to characterize the emission sources as well as the evolution of particle size distribution in the proximity of these emission points. In this study, the authors evaluated the particle concentration and size distribution at a downwind receptor site of a linear (a major highway) and point (waste incinerator plant) source in an area characterized by high anthropic environmental impact. The particle emissions of the incinerator under examination were characterized by using a Scanning Mobility Particle Sizer® (SMPS), an Aerodynamic Particle Sizer® (APS) Spectrometer, a Rotating Disk Thermodiluter and a Thermal Conditioner (Matter Engineering AG). As regards the linear source, concentrations were determined at increasing distances from the most important Italian road, the A1 highway. Particle number, surface and mass exponentially decreases away from the freeway, whereas particle number concentration measured at 400 m downwind from the freeway is indistinguishable from upwind background concentration. Annual mean values of $8.6x10^3 \pm 3.7x10^2$ particle cm^{-3} and 31.1 ± 9.0 µg m^{-3} were found for particle number and PM_{10} concentration, typical of a rural site. The particle apportionment and exposure assessment in respect of linear and point sources for ultrafine particles represent the major novelty of the present paper. The study here presented could be very important in developing appropriate management and control strategies for air quality in areas characterized by high anthropic pressure and to perform exposure assessment for populations involved.

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1. Introduction

Several epidemiological studies correlate particulate matter to negative health effects such as cardiovascular problems, increase of mortality and morbidity and negative effects on human breathing (Dockery et al., 1993; Dockery et al., 1996; Cheng, 2003; Kreyling et al., 2006). In particular, in the last years, toxicologists have extended their attention on fine ($PM_{2.5}$ – particulate matter with aerodynamic diameter less than 2.5 µm) (Pope and Dockery, 2006) and ultrafine particles (UFPs, often defined as smaller than 0.1 µm), but there is no consensus in the scientific community on which particle property has the worst effects on human health (particle number, mass, surface area concentration or chemical composition). Emission inventories suggest that the highest contribution to the fine and ultrafine particles come from anthropogenic activities, namely from emissions of industrial combustion processes and traffic-related emissions (Schauer et al., 1996; Shi et al., 1999; Airborne Particles Expert Group, 1999; EPA, 2000; Cass et al., 2000; Harrison et al., 2000; Hitchins et al., 2000). In urban, rural or industrial areas the literature on systematic monitoring of ultrafine particles is not as extended as that for PM_{10} (particulate matter with aerodynamic diameter less than 10 µm) and $PM_{2.5}$, mainly because current regulation does not target the concentration of these particles (Fine et al., 2004).

On the contrary, the temporal concentration and size distribution of particles in proximity of linear sources such as highways or main roads in urban areas have been deeply investigated (Hitchins et al., 2000; Zhu et al., 2002a; Zhu et al., 2002b; Kittelson et al., 2004; Buonanno et al., 2009a). In urban areas there are generally two main contributors to the particle concentrations: primary or direct emissions from particle sources and secondary particulate matter formed by photochemical or physical processes in the atmosphere. The aerosol at a downwind receptor location will be then constituted by particles emitted from local sources as well as particles emitted or formed far upwind and aged during transport (Trier, 1997; Morawska et al., 1998; Hughes et al., 1998; Harrison et al., 2000; Woo et al., 2001), with diurnal concentration profiles that generally match those of local vehicular sources. However, the majority of these studies regarded periods ranging from a few weeks to a few months.

In the case of industrial areas, there is a lack of information on ultrafine particle monitoring even if the stack emissions were analyzed for several energy production plants (Schmatloch, 2000; Ohlstrom et al., 2000; Gaegauf et al., 2001; Maguhn et al., 2003; Chang et al., 2004; Wierzbicka et al., 2005; Buonanno et al., 2009b). The emission factor in terms of particle number highly depends on the flue gas treatment (Buonanno et al., 2009b). In the waste field, incineration represents a favorable technique for reducing the volume of waste streams and recovering its energy content for generating electricity and district heating (UBA, 2001; TWGComments, 2003). Incineration is used as a treatment for a very broad range of wastes even if it generally represents only one part of a complex waste treatment system that altogether provides for the overall management of the broad range of wastes that arise in the society. The incineration sector has undergone rapid technological development over the last 10 – 15 years, due to

specific legislation applied to industry that has obliged several European countries to reduce toxic emissions from municipal waste incinerators (MWIs) (European Commission, 2006). However, the fine and ultrafine particle stack emission has not yet been fully characterized. Because of this, MWIs represent a rather interesting subject of investigation. In Western countries there is a strong debate on the emission of ultrafine particles at the stack of waste-to-energy plants, although MWIs surely represent only a minor source of anthropogenic aerosol emission compared to fossil fuel power plants and vehicle emissions (Airborne Particles Expert Group, 1999; EPA, 2000; Cass et al., 2000).

The present paper intends to evaluate the seasonal concentrations and size distributions of particles in terms of number and mass in the area of San Vittore del Lazio (Italy) characterized by high anthropic pressure (environmental impact due to anthropic activities). The authors consider this an area of extreme importance in order to analyze the environmental impact in terms of ultrafine particles of a waste-to-energy plant (point source) compared to a highway (linear source). The particulate matter (PM) data presented in this article were produced over a 12-month period at a sole location within the above-mentioned area. The location can be considered a downwind receptor site. The sources and formation mechanisms of PM in this site are also of particular interest. In fact, this site is influenced by local primary sources, the advection of primary but aged particulate emissions from areas situated upwind, and the formation of secondary aerosols in the atmosphere.

2. Experimental

2.1. Site description

The site selected for the present study (41°26′19″N–13°53′46″E) is located in the area of San Vittore del Lazio, in the center of Italy. Two predominant PM sources were considered: (i) a waste-to-energy plant (point source) and (ii) the A1 "Autostrada del Sole" highway (linear source) (see Figure 1).

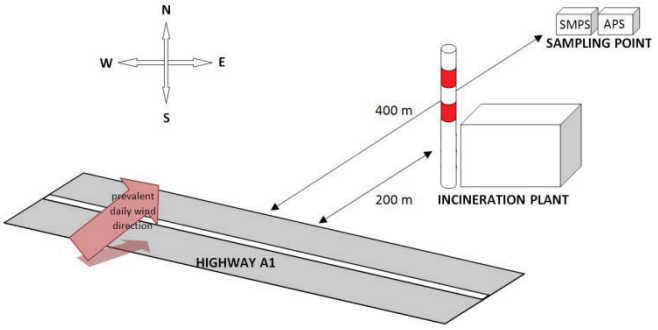

Figure 1. *Location of the sampling site in the San Vittore del Lazio area.*

The main purpose of the plant is the waste-to-energy treatment of Refuse Derived Fuel (RDF), sorted from municipal solid waste (MSW) with the non-combustible fraction removed. It presents a high heating value fuel suitable to produce electricity. The incinerator consists of the following main sections: (i) the waste delivery area, where the delivery trucks arrive in order to dump the waste into the bunker, (ii) the combustion and heat recovery section, consisting of a moving grate incinerator and an additional burner system to maintain the required minimum temperature (850 °C) (Directive 2000/76/EC), (iii) the power generation section, constituted by a condensing turbine unit directly coupled to the generator, and (iv) the flue-gas treatment section (semi-dry process) made up of a selective non-catalytic reduction (SNCR), a spray absorber system (lime milk and powdered activated carbon) and a fabric filter. In the SNCR process, NO_X in the flue gas is reduced to N_2 by reaction with urea $CO(NH_2)_2$ at high temperatures (900 – 1 000 °C), avoiding the use of a catalyst. The main technical data of the plant are summarized in Table 1.

The A1 highway presents a W-E direction, six lanes (plus two emergency lanes), three west-bound and three east-bound. It is approximately 26 m wide (excluding the emergency lines). As regards to the traffic density, it can be divided into two types in terms of weekdays (Buonanno et al., 2009a). The daily traffic is predominant during the weekdays whereas it increases significantly just before the weekend. The estimated traffic conditions can be summarized as: (a) daily traffic, with a traffic density of 53 ± 15 vehicle min^{-1} passing the sampling site in both directions (23% heavy-duty vehicles), (b) weekly traffic, with a traffic density of 95 ± 12 vehicle min^{-1} (12% heavy-duty vehicles). Average vehicle speed ranged from 25 to 40 m s^{-1} (90 – 140 km h^{-1}) (Buonanno et al., 2009a).

Particles were sampled over a 12-month period at one location within the above-mentioned area, at a distance of about 200 m from the incinerator and 400 m from the A1 highway (Figure 1). The sampling location can be considered a downwind receptor site for the area. In fact, the site is characterized by a daily SW predominant wind direction (with a more constant direction in the summer period) that carries the highway vehicular and stack incinerator emissions directly to the sampling location (Figure 2), whereas at night, an inversion during the predominant wind is observed both in winter and in summer.

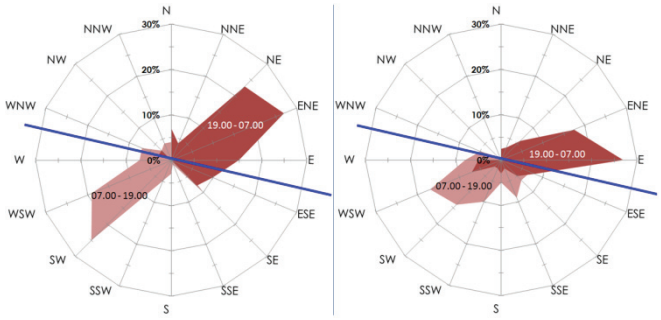

Figure 2. *Wind direction frequency in the day (7:00 - 19:00) and night (19:00 - 7:00) during the (a) winter and (b) summer season. The blue line represents the A1 highway.*

The incinerator continuously operated during the winter (from November to February) and summer seasons (from June to September). The mean winter temperature varies between 3 °C and 12 °C whereas the mean summer temperature is comprised between 17 °C and 30 °C.

Finally, the location selected can also be considered as ideal because of the absence of important upwind residential areas. Then, the upwind sources could be expected to have little or no appreciable effect on the sampling site.

2.2 Instrumentation and methodology

The experimental apparatus is constituted by:

- a Scanning Mobility Particle Sizer (SMPS 3936, TSI Inc.) composed of a Condensation Particle Counter (CPC 3775) and an electrostatic classifier (EC 3080), able to detect particles with dimensions between 6 and 1 000 nm and particle number concentrations from 2 to $1x10^7$ particle cm^{-3};

Table 1. *Parameters and performance of the incineration plant*

Parameter	Value
Refuse Derived Fuel capacity	12x10^3 kg h^{-1} with a low heat value greater than 15 MJ kg^{-1}
Plant annual availability	8 000 h
Gross electrical power	11.7 MW
Solid residuals	Bottom ashes: 14x10^6 kg year^{-1}
	Fly ashes: 5 x10^6 kg year^{-1}
Reagent consumption in flue gas treatment	Urea: 160 m^3 year^{-1}
	Powder activated carbons: 80 x10^3 kg year^{-1}
	Calcium oxide CaO: 1.6 x10^6 kg year^{-1}
Stack emission characteristics	Flowrate: 100 000 m^3 h^{-1}
	Temperature: 140 °C
	Height: 50 m
	Exhaust speed at the stack: 12 m s^{-1}
	Annual mean NO_x concentration at the stack: 123 mg Nm^{-3}
	Annual mean SO_2 concentration at the stack: 7.9 mg Nm^{-3}
	Annual mean dust concentration at the stack: 0.3 mg Nm^{-3}

- an Aerodynamic Particle Sizer (APS 3321, TSI, Inc.) able to evaluate the aerodynamic diameter of particles in the range of 0.5 – 20 μm measuring the time of flight (TOF) with a 52 channels resolution and particle number concentration starting from 10^{-3} to 10^3 particle cm^{-3};
- a Rotating Disk Thermodiluter (Matter Engineering AG) and a Thermal Conditioner (Matter Engineering AG) together with a Scanning Mobility Particle Sizer (SMPS, TSI Inc.) were used to characterize the particle number distributions and total concentrations of the particles emitted by the waste incinerator. Details of the instrumentation are reported in (Hueglin et al., 1997);
- a weather station (Davis Vantage Pro) to estimate the main climatic parameters (temperature, relative humidity, atmospheric pressure, rainfall, wind direction and velocity, and solar irradiation) starting from 2004.

Using the SMPS-APS system, the particle number, surface area and volume particle distribution in the 10 nm–10 μm aerodynamic diameter range can be determined. The measurements conducted with the SMPS-APS system have been obtained every 30 minutes with sampling time of 240 s and every value represents the arithmetic average of three measurements. Particle mass concentration has been determined by evaluating a constant value of the particle density equal to 1.7 g cm^{-3} (Buonanno et al., 2009c) and on the basis of an algorithm well described in Fine et al. (2004) and Sioutas et al. (1999).

As regards to the characterization of the particles emitted by the point source, experimental campaigns were conducted in November 2007, June 2008, and October 2009 at the stack of the incinerator in order to estimate the emissions of the waste incinerator in terms of total particle number concentration. The design of the sampling and dilution system determines largely what is measured later. It serves to reduce the concentration in the raw exhaust to a concentration which can be handled by the measurement system and to control the condensation and nucleation processes (Burtscher, 2005). Cooling the exhaust with no or moderate dilution will cause a strong nucleation/condensation.

For this purpose a custom-made sampling line was designed and constructed. It was constituted by the following parts: (i) 650 mm long stainless steel probe, (ii) Zambelli probe electric heater, (iii) Matter Engineering AG Rotating disk model MD19-2E and Air Supply Thermal Conditioner Aset 15-1.

The sampling probe (stainless steel), all sampling lines, and the first part of the dilution system were heated with an electric heater (Zambelli) to temperatures of about 150 °C to avoid condensation effects in the probe. Furthermore, the coupling flange and the final part of the probe were insulated with glass wool. Finally, the sampled combustion aerosol was rapidly diluted by a factor of 25 with clean dry air.

The sizing accuracy of the instrumentation was verified in the TSI laboratory in High Wycombe, UK a few days before the start of the experimental campaign, by means of monodisperse polystyrene latex spheres. The post-processing analysis was performed through the Aerosol Instrument Manager® and Data Merge® (TSI Inc., St. Paul, MN) software, together with the authors' custom-made subroutines. The uncertainty budgets in measuring the particle number and mass distributions and concentrations in terms of absolute expanded uncertainty (k=2, level of confidence 95%) (ISO/IEC Guide 98-3, 2008) were determined through the model reported in Buonanno et al. (2009c).

3. Results and Discussion

3.1. Characterization of the waste incinerator emissions in terms of ultrafine particles

The measurements were performed from the stack at a height of 35 m. The operational conditions for the 15th of October are reported as an example in Table 2. The data, obtained on the basis of semi-hourly values, show a very steady combustion conditions.

Table 2. *Operative mean conditions and corresponding standard deviation for the main parameters during the October 2009 experimental campaign*

Parameters	Mean value	Standard deviation
Normalized flow rate (Nm^3 h^{-1})	98.3x10^3	1.7x10^3
Stack temperature (°C)	135	4.1
Combustion chamber temperature (°C)	991	9.6
Relative humidity (%)	15.3	1.0
O_2 in dry flue gas (%)	10.7	0.3
SO_2 (mg Nm^{-3}]	8.2	1.2
NO_x (mg Nm^{-3})	115.2	8.7
CO (mg Nm^{-3})	5.2	1.7
Dust (mg Nm^{-3})	0.68	0.11
HCl (mg Nm^{-3})	4.3	0.3

In Figure 3, the particle number distribution (based on the average value of tests carried out during October 2009) versus the aerodynamic diameter is reported. In particular, the distribution presents a mode at about 90 nm. The obtained distribution and the corresponding mode value is in good agreement with the experi-

mental results reported by Maguhn et al. (2003) analyzing the stack gas of a municipal waste incineration plant (23 MW) at a stack temperature of 80 °C and in Buonanno et al. (2009b) where the stack particle emissions (at 170 °C) of a waste-to-energy plant (12 MW) were monitored. The total particle number concentration varies from $1x10^2$ particle cm^{-3} to $1x10^3$ particle cm^{-3}. In the plant analyzed, the presence of the fabric filter in a semi-dry flue gas treatment allows an optimum performance in reducing not only the total particle mass, but also the total particle number emission. For this purpose, the particle number distribution and concentration were also measured before the fabric filter, showing an efficiency of 99.995% in terms of total particle number concentration.

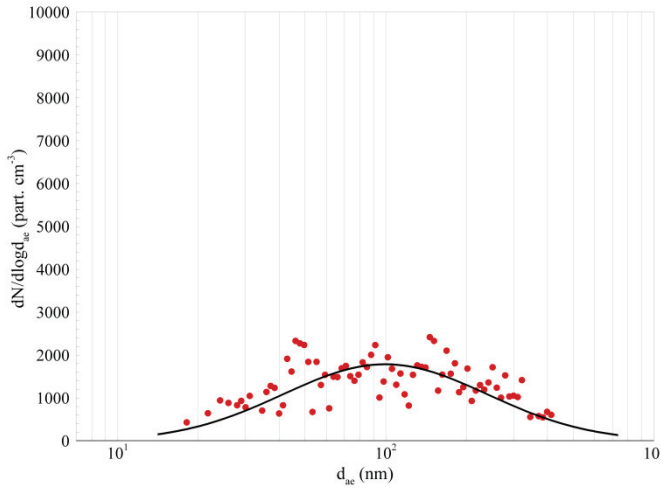

Figure 3. *Particle number distribution at the stack of the incinerator plant.*

3.2. Characterization of the A1 highway in terms of ultrafine particles

Details of the experimental campaign are reported in Buonanno et al. (2009a). Figure 4 shows the ultrafine size distribution decay measured at different sampling locations (Z-axis) near the A1 highway in the case of weekly traffic. The X-axis represents the aerodynamic diameter on a logarithmic scale whereas the Y-axis indicates the particle number concentration. Only one dominant particle mode was observed for all sampling locations in the case of weekly traffic. At 30 m downwind, this mode occurred around 7 nm with a modal particle number concentration of $6.0x10^5$ particle cm^{-3}. This mode persisted at distances up to 400 m without shifting to a larger size. The ultrafine particle concentrations measured at 400 m downwind from the A1 highway were still different from the background concentrations. The obtained results agree very well with those reported by Zhu et al. (2004) and referred to the 405 freeway (with a dominant gasoline vehicle emission, in winter with a mean temperature of 23.2 °C). The differences between the ultrafine particle concentrations measured at 400 m downwind from the A1 highway with respect to the background concentrations (in Zhu et al. (2004) they were compared at 300 m) can be roughly explained with the higher mean wind speed value (3.1 m s^{-1} in this study compared to 1.3 m s^{-1} in Zhu et al. (2004)). The higher the wind speed from the highway towards the sampling points, the greater the distance from the highway influenced by traffic emissions (Hitchins et al., 2000). In Figure 4 the particle number size distribution emitted by the incinerator is also reported. With respect to the contribution of the linear source, the emission of the waste incinerator has to be considered as negligible.

3.3. Experimental results at the downwind receptor site

PM_{10}, $PM_{2.5}$ and particle size distribution (PSD) measurements were obtained through the SMPS-APS system. The relationship between particle mass and number aerosol particle concentrations typically shows very low correlation. In the present study, a good correlation was found when data are divided by weeks as shown in Figure 5. The high correlation can be related to the presence of a dominant source that is able to influence both the particle number and mass concentrations. The correlation during the summer period is higher than the one referred to the winter time due to a more frequent and constant wind from the SW direction, from the main sources to the downwind receptor site. The average distributions are reported in Figure 6. The particle number distribution presents a mode equal to 168±34 nm and to 138±28 nm in the winter and summer, respectively. As regards to the particle mass distribution, it shows a bimodal shape, with the first mode in the accumulation mode size range equal to 372±74 nm and constant during the year. Especially during the summer season, the modality of the particle size distribution (aerosol characteristic that can be associated with the aerosol mechanism formation) is typical of a traffic-influenced aerosol (Morawska et al., 1999) even if the total particle number and mass concentration are very low, typical of a rural site (see Table 3). The summer particle number size distribution reported in Figure 6b shows, with respect to the winter data (Figure 6a), higher concentrations of particles in the 10 – 80 nm range and lower concentrations in the other part of the range. Once again, the presence of a "fresh" aerosol from a local particle source can be related to the more frequent and constant wind speed and direction during the summer season from the main sources to the downwind receptor site. Table 3 reports the total particle number and mass concentrations and the corresponding mode values by considering the annual, winter and summer period. The annual mean total concentration values were $8.6x10^3$±$3.7x10^2$ particle cm^{-3} and 31.1±9.0 µg m^{-3} in terms of total particle number and PM_{10} concentration, respectively. They represent very low values compared to the typical Italian values, characteristic of a site with a modest contribution of anthropogenic sources. The highest values are found during the winter period, characterized by a thermal inversion condition of the atmosphere with an increasing mixing height in the afternoon.

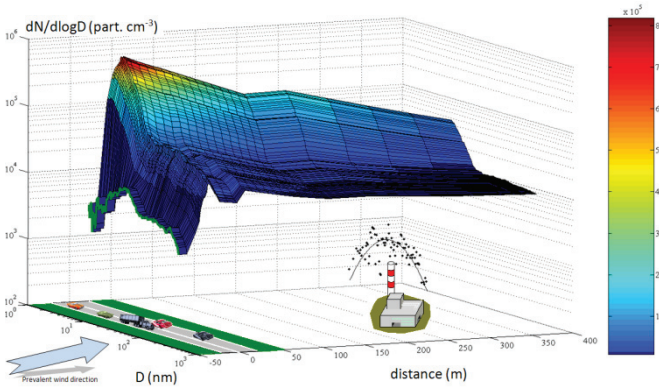

Figure 4. *Particle number distribution in the San Vittore del Lazio area. The green line at upwind position in respect to the highway represents the background particle number distribution.*

In order to develop an ultrafine particle apportionment, in Figure 7 the total particle number and mass (PM_{10}) concentrations are reported as a function of the day of the week during the summer and winter seasons. In both seasons the concentrations during the weekends are much lower compared to the weekday

concentrations. For example, in summer, the mean weekend concentrations are 44% and 60% lower than the mean weekday concentrations for particle number and mass, respectively. These differences highlight the main influence of a source like the A1 highway whose emissions are predominant during the day, as further confirmed in Figure 8 where the daily particle mass and number concentration trends in the winter and summer seasons are reported. In both seasons, traffic emissions can be considered as the main source because of the presence of a double particle mass and number concentration peak in the morning and in the evening. During these periods (8:00 – 10.00 am and 4:00 – 6.00 pm), the particle number is very high showing the presence of fresh particles coming from the linear source that has to be considered as a local source with respect to the sampling point. In summer at 6.00 pm, the peak referred to PM_{10} is higher compared to the winter one, because of, once again, a more constant wind intensity and direction.

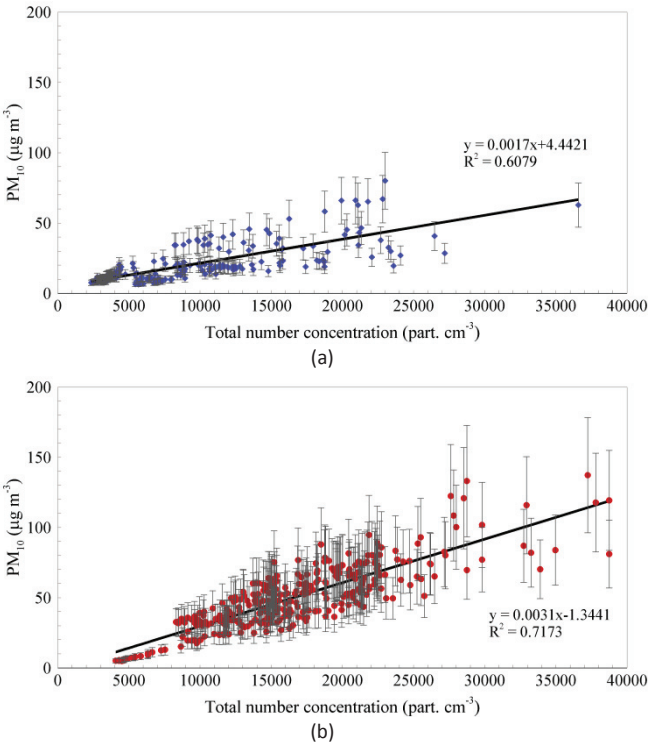

Figure 5. *Correlation between PM_{10} and particle number concentration from (a) the 7th up to 13th January (winter time) and from (b) the 5th up to 11th September (summer time).*

In Figure 9a the mean PSD winter evolution during weekdays and weekend in the sampling site is shown. The weekday concentrations are higher than weekend ones. During the weekdays, at 9:00 am and 6:00 pm there is a significant amount of fresh particles coming from the highway. The corresponding mode in the nuclei mode size range is equal to about 40 - 50 nm, typical of a site located 400 m downwind on a road characterized by a high traffic volume (Hitchins et al., 2000; Zhu et al. 2002a; Zhu et al. 2002b; Kittelson et al., 2004; Buonanno et al., 2009a). At 2:00 pm the total concentration is diminished and the PSD is bimodal with a nucleation mode of about 30 nm and the usual peak in proximity of 150 nm. The total concentrations of the corresponding PSD during the weekend are lower with a unimodal distribution typical of aged particles. During the summer season (Figure 9b), the above-mentioned considerations are confirmed even if a higher difference between weekday and weekend values can be found due to the intensity and more constant wind direction from the sources to the downwind site receptor.

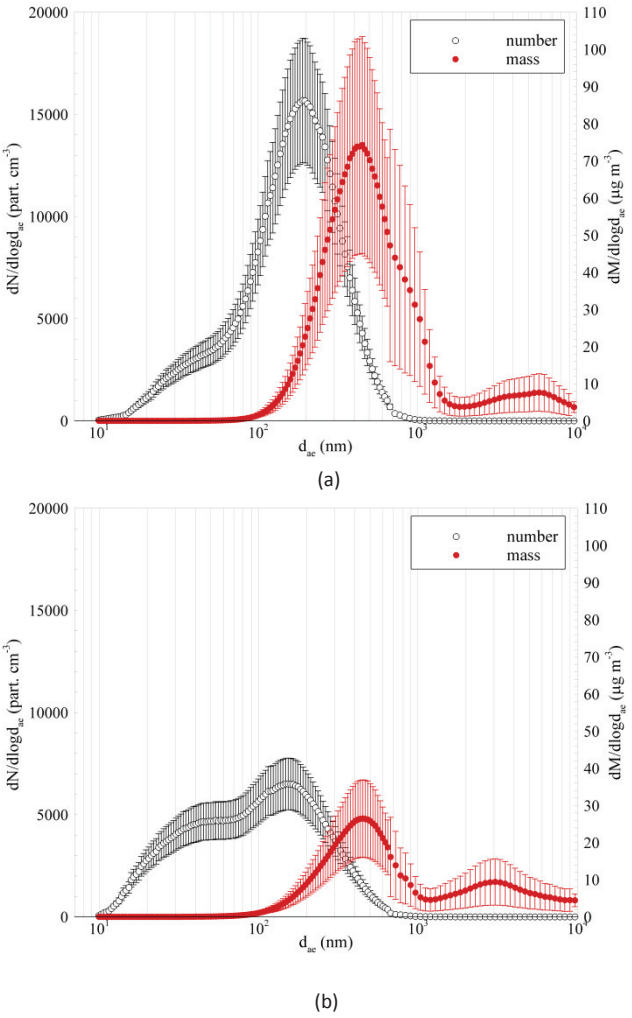

Figure 6. *Seasonal particle number and mass distributions in winter (a) and summer (b) at the area of San Vittore del Lazio (Italy). The uncertainty budget was determined through the model reported in Buonanno et al. (2009c).*

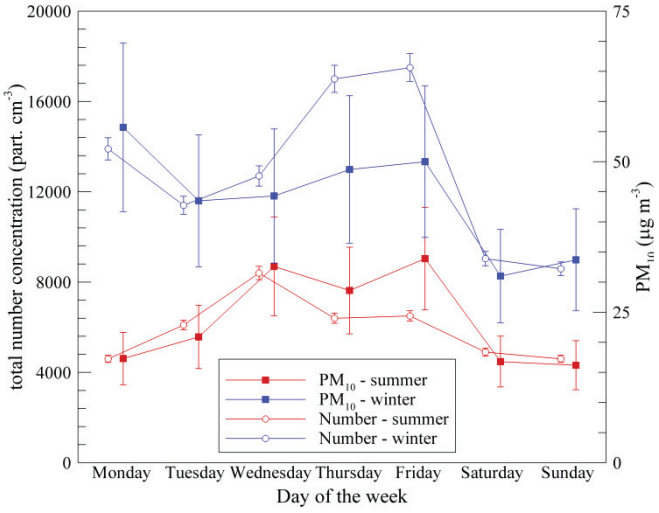

Figure 7. *Total particle mass (PM_{10}) and number daily concentrations in winter and summer seasons. The uncertainties were evaluated through the model proposed in Buonanno et al. (2009c).*

Table 3. *Total particle number and mass concentrations and corresponding mode values during the annual, winter and summer periods. The uncertainties were evaluated through the model proposed by Buonanno et al. (2009c)*

	Winter mean value	Summer mean value	Annual mean value
Total particle number concentration (particle cm^{-3})	$1.1 \times 10^4 \pm 5.0 \times 10^2$	$6.2 \times 10^3 \pm 2.1 \times 10^2$	$8.6 \times 10^3 \pm 3.7 \times 10^2$
Particle umber distribution mode (nm)	168±34	138±28	153±31
PM_{10} ($\mu g\ m^{-3}$)	38.4±10.7	23.9±7.4	31.1±9.0
Particle mass distribution mode (nm) (accumulation mode size range)	372±74	372±74	372±74
Particle mass distribution mode (nm) (coarse mode size range)	4 068	2 642	3 355

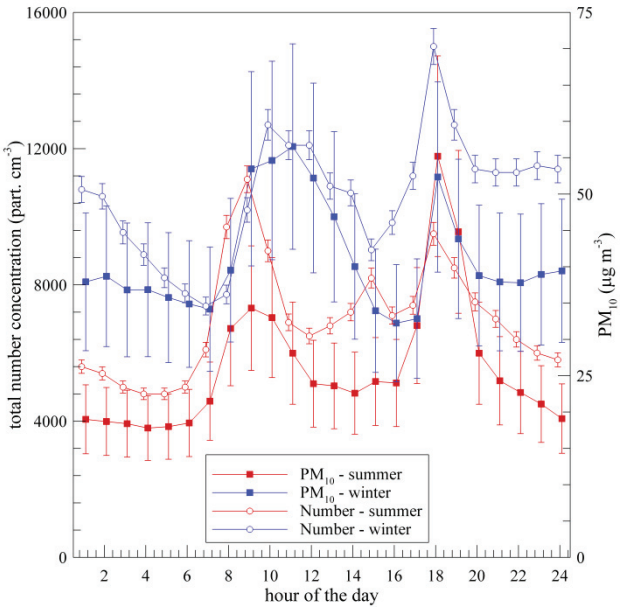

Figure 8. *Hourly particle mass and number concentration trends in winter and summer seasons. The uncertainties were evaluated through the model proposed in Buonanno et al. (2009c).*

4. Conclusions

In the present study, a 12-month period experimental analysis was carried out in the area of San Vittore del Lazio (Italy) characterized by high anthropic pressure to evaluate the seasonal concentrations and size distributions of particles in terms of particle number and mass, and also in order to apportion ultrafine particle emissions from a linear (major highway) and a point source (waste incinerator). Waste incineration represents a favorable technique for reducing the waste volume. However, in the past, municipal waste incinerators (MWIs) had a bad reputation due to the emission of toxic combustion byproducts. Consequently, the risk perception of the people living near waste incinerators is very high even if in Western countries waste incineration has nowadays to be considered a relatively clean process. The particle apportionment and exposure assessment in respect of linear and point sources for ultrafine particles represent the major novelty of the present paper. The study here presented could be important for developing appropriate management and control strategies for the air quality in the surroundings of waste incinerators and to perform exposure assessment for populations involved.

Through a SMPS-APS system, particle number and mass concentrations were measured at a downwind receptor site whose results are typical of rural sites ($8.6 \times 10^3 \pm 3.7 \times 10^2$ particle cm^{-3} and 31.1±9.0 $\mu g\ m^{-3}$ for total particle number and mass concentration, respectively), with a modality typical of traffic-influenced sites.

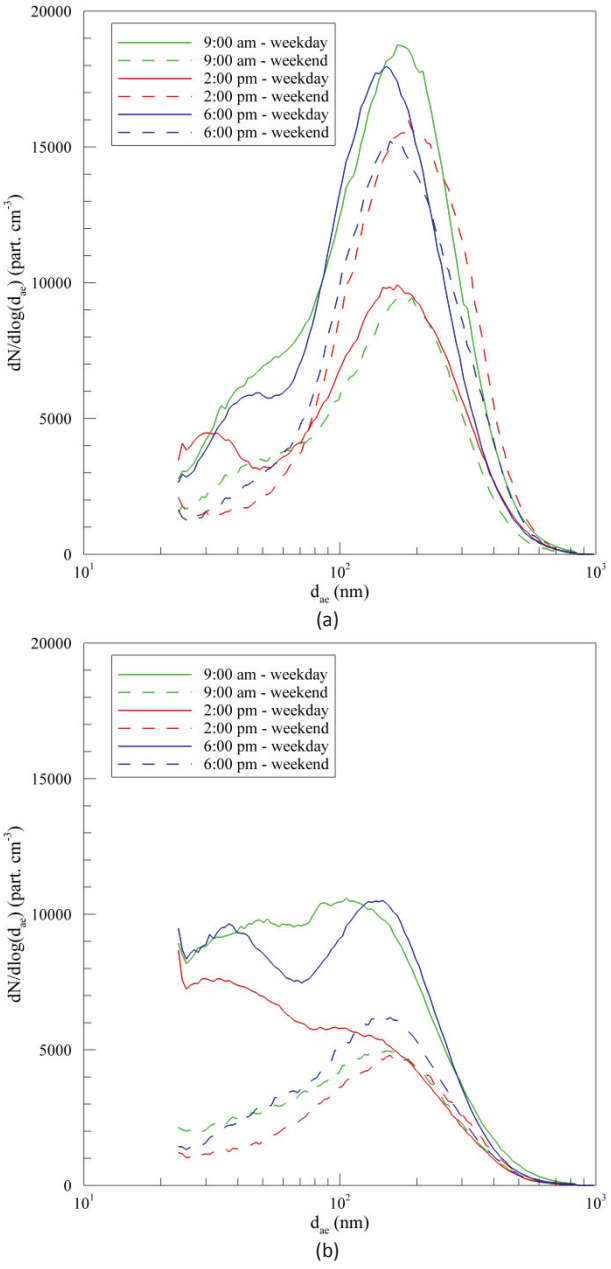

Figure 9. *Particle number distributions at different time in winter (a) and summer (b). The uncertainties were evaluated through the model proposed in Buonanno et al. (2009c).*

The effect of the incineration plant could only be restricted to the emission of precursor gases for secondary particle formation and it is negligible with respect to the A1 highway. In fact, the main influence of a source like the A1 highway whose emissions are predominant during the day can be observed through the temporal data series (on weekly and daily basis) in terms of particle number and mass concentrations. The particle size distributions (PSDs) of the highway were similar to PSDs obtained by other researchers having similar highway traffic conditions. The daily PSD evolution here analyzed, shows the main contribution of the highway also in terms of secondary particles due to the fresh particles coming from the highway.

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