Charged particles and cluster ions produced during cooking activities

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Abstract

In this paper we show, for the first time, the electric charge of particles generated during cooking activities and thus extending the interest on particle charging characterization to indoor micro-environments, so far essentially focused on outdoors.

Particle number, together with positive and negative cluster ion concentrations were monitored using a condensation particle counter and two air ion counters, respectively, during different cooking events. Positively-charged particle distribution fractions during gas combustion, bacon grilling, and eggplant grilling events were measured by two Scanning Mobility Particle Sizer spectrometers, used with and without a neutralizer. Finally, a Tandem Differential Mobility Analyzer was used to measure the charge specific particle distributions of bacon and eggplant grilling experiments, selecting particles of 30, 50, 80 and 100 nm in mobility diameter.

The total fraction of positively-charged particles was 4.0%, 7.9%, and 5.6% for gas combustion, bacon grilling, and eggplant grilling events, respectively.

Keywords: cooking-generated aerosol, ultrafine particles, particle charge, ion concentration, TDMA, gas combustion.

1. Introduction

A number of epidemiological and toxicological studies have shown a positive link between inhaled ultrafine particles (UFPs, particles with diameter smaller than 100 nm) and human health (Sayes et al., 2007). These adverse effects are mainly due to the ability of UFPs to penetrate into the human respiratory system, depositing in the deepest regions of the lung, while carrying a number of toxic compounds (International Commission on Radiological Protection, 1994).

People are exposed to airborne particles from a range of sources (Morawska et al., 2008; See and Balasubramanian, 2006) leading to large doses associated with every type of lifestyle. Personal dose is a function of the particle concentration that people are exposed to in a given microenvironment (Buonanno et al., 2011a; Buonanno et al., 2012c) and therefore, an accurate evaluation of the dose can only be made if particle concentration levels in that microenvironment are known. Consequently, several studies have been performed to characterize both indoor and outdoor microenvironments in terms of particle number, surface area and mass distributions, and total concentrations (Buonanno et al., 2012a; Buonanno et al., 2013; Buonanno et al., 2010; Kaur et al., 2005; Morawska et al., 2008; See and Balasubramanian, 2006).

In addition to the above factors, the dose received by humans is strongly related to particle deposition efficiencies in the various regions of the lung. Nowadays, the evaluation of dose is generally based on inhaled particle deposited fraction data provided by the International Commission on Radiological Protection (1994), as a function of the inhalation rate, type of activity performed and region of the lung. However, deposition fractions do not take into account factors such as the electric charge on the particles. Several studies have shown that the presence of charge on particles may increase the deposition rate in the lungs (Chan et al., 1978; Chan and Yu, 1982; Majid et al., 2011; Melandri et al., 1983), with one study suggesting that the deposition rates may be enhanced by factors as high as three or five (Cohen et al., 1998). Therefore, ignoring the charge on these particles may significantly underestimate the actual dose received by subjects. In fact, in the alveolar and tracheobronchial regions of the lungs, charge-neutral particles in the range 100-200 nm are deposited with a lower efficiency (International Commission on Radiological Protection, 1994), nonetheless if these particles present an elevated charge state (highly charged, e.g. due to their large surface area) their deposition is expected to increase, leading to higher total particle doses.

1.1 Cooking-generated particle characterization: state-of-the-art

In order to perform an overall assessment of personal exposure to particles, particular attention should be paid to indoor microenvironments where people spend the majority of their time (80-90%). Among other indoor sources (e.g. candles, incense and other aesthetic products (Stabile et al., 2012)), the main source of indoor UFPs is cooking activities. In fact, several studies have been performed to characterize cooking-generated particles, including derivation of their emission rates and size distributions (Buonanno et al., 2009; See and Balasubramanian, 2006). Particle number emission rates in the range 10^{10}-10^{13} part. min^{-1} were measured and found to vary according to cooking method (grilling, frying), type of food (fat-rich, vegetable foods), cooking temperature and type of cooking oil used (Buonanno et al., 2009; Wallace et al., 2008). Such strong emissions can lead to high number concentrations (>1×10^5 part. cm^{-3}) of cooking-generated particles in indoor environments and these are likely to remain airborne long after the cooking activity had ceased (Burtscher et al., 1986). In fact, cooking/eating time was found to be the main contributing activity in the personal daily dose of children (Buonanno et al., 2012b); Ko et al. (2000) recognized food cooking as the main contributor to lung cancer for nonsmoker Chinese.

The chemical properties of the aerosols produced during cooking activities have also been reported in scientific literature. For example, Elmore et al. (2000) and Elmore et al. (2004) identified several volatile compounds produced during meat grilling, including many hydrocarbons, alcohols, ketones and aldehydes. Similarly, Byrne et al. (2002) identified twenty-six volatile components, including aliphatic alkanes, saturated and unsaturated aldehydes, ketones and 1-octen-3-ol, produced during the cooking of chicken patties.

Moreover, cooking-generated particle volatility was investigated by Buonanno et al. (2011b), who performed particle number distribution measurements after aerosol thermal conditioning at different temperatures. They showed a significant reduction in particle number concentration for vegetable foods compared to fatty foods, recognizing that the presence of a solid core was likely to result in the partial synthesis and degradation of fatty acids into aldehydes and ketones.

Finally, Buonanno et al. (2009) performed morphological characterization of the UFPs collected during grilling activities using a TEM and documented aggregate structures showing an average primary particle diameter of about 30 nm and a fractal dimension lower than 2.0.

In summary, while in-depth physical, chemical and morphological characterization of cooking-generated particles has been carried out in the past decades, analysis of the electrical charge of these particles has not yet been performed by the scientific community.

1.2 Electrically charged particles

The charge characteristics of particles emitted by motor vehicle engines have been measured (Maricq, 2006) and high concentrations of charged particles near trafficked roads have been observed (Lee et al., 2012). However, there is a lack of knowledge regarding the charge characteristics of cooking-generated aerosol particles. A better knowledge of particle charge characteristics is a key aspect in aerosol measurement field since the operating principles of instruments measuring particle size distributions and concentrations (e.g. mobility analyzers, lung-deposited surface area monitors) is based on the knowledge of the particle charging efficiency (charge distribution; Kinney et al. (1991)). Anyway, freshly-generated particles may present an initial (pre-existing) charge distribution which is not easily neutralized by instruments using unipolar diffusion chargers (Kaminski et al., 2013; Leskinen et al., 2012; Qi et al., 2009) causing possible aerosol mischarging and related wrong measurements.

Airborne particles are charged due to the attachment of “small ions” (singly charged molecular clusters smaller than about 2 nm in size, also known as ‘cluster ions’) (Hirsikko, 2011). Therefore, cluster ion concentration is one of the controlling parameters in the particle charging process.

Atmospheric positive and negative ion concentrations have been measured in the range 200-2500 cm^{-3} (Hirsikko, 2011). In particular, concentrations of 300-400 cm^{-3} typically occur under stable atmospheric conditions, whereas natural (e.g. waterfalls, rainfalls (Hirsikko, 2011; Laakso, 2007)) and anthropogenic sources (e.g. powerlines, motor vehicles, trafficked roads (Jayaratne, 2011; Jayaratne et al., 2008; Jayaratne et al., 2010; Ling et al., 2010; Maricq, 2005)) can increase concentrations by up to a few thousand cm^{-3}.

In order to assess the interaction rate of cluster ions with airborne particles, the electrical charge of the particles has been investigated by performing particle charge distribution measurements using a Tandem Differential Mobility Analyzer (TDMA) system (Lee et al., 2012; Maricq, 2006). Under stable conditions and in the presence of symmetric positive and negative small ion concentrations, particles are charged in accordance with the equilibrium charge distribution. Combustion processes are known to emit both positive and negative ions at approximately the same rate (Maricq, 2006, 2008), due to the fact that the charging production processes involved are bipolar (chemi-ionization and hydrocarbon flames; Wright (2007)). Burtscher et al. (1986) showed that charge distribution depends strongly on the combustion material. Therefore, charging characteristics of particles emitted by the combustion of different combustible substances should also be analyzed. The particle charge distribution also depends on flame temperature, however, when the aerosol leaves the flame (coagulating further), its charge state decreases to the Boltzmann distribution at room temperature in few seconds (Maricq, 2008).

1.3 Aims of the work

In the present paper, the results of an experimental campaign aimed to evaluate whether cooking activities produce ions and charged particles are reported. Particle charge distributions and ion concentrations in indoor air during cooking activities are reported for the very first time thus improving the understanding of cooking-generated aerosols. Grilling experiments were preferred as they are among the main particle-emitting cooking activities (Buonanno et al., 2009) and also represent easily repeatable experiments since no other parameters affecting the particle emission rate have to be taken into account (e.g. oil type and/or oil level in the pan during frying). The indoor particle and cluster ion concentration measurements were carried out using condensation particle counters and ion counters, respectively, whereas particle charge distributions were measured using a TDMA system.

2. Materials and methods

2.1 Site description

Measurements were performed during June-September 2012 at the European Accredited (EA) Laboratory of Industrial Measurements (LAMI), University of Cassino and Southern Lazio, Italy. The room had an area of about 50 m^2 (150 m^3) and thermo-hygrometric conditions were continuously monitored, in order to maintain temperature and relative humidity values of 20±1 °C and 50±10%, respectively. A portable kitchen unit with gas stoves (mixture of butane and propane) was set-up inside the room. The ventilation conditions were held constant during the tests (doors and windows closed with mechanical ventilation in operation) and the air exchange rate (AER), evaluated on the basis of the CO_2 decay curve (He et al., 2004) using a TSI Model 7515 IAQ-CALC, was measured to be 0.3±0.1 h^{-1}.

2.2 Experimental apparatus

To perform cluster ion concentration, particle concentration, particle distribution and charge distribution measurements, the following instruments were used:

- two battery-operated Air Ion Counters (AICs, Alphalab Inc.) to simultaneously measure positive and negative cluster ion concentrations up to 2×10^{6} ions cm^{-3} with a resolution of 10 ions cm^{-3};
- two Condensation Particle Counters (CPC 3775, TSI Inc.) to measure total particle number concentrations up to 1×10^{7} part. cm^{-3} of particle size down to 4 nm;
- two Electrostatic Classifiers (ECs 3080, TSI Inc.) to classify particles according to their electrical mobility. These include a radioactive Aerosol Neutralizer (3077A, TSI Inc.) to impose a bipolar charge distribution on the aerosol, and a Long Differential Mobility Analyzer (DMA 3081, TSI Inc.) where positively-charged particles of the required electrical mobility (i.e. mobility diameter) are classified in a negative high voltage electric field generated in the classification region between an inner controlled negative voltage cylindrical rod and an electrically grounded outer cylinder.

ECs and CPCs were also used in tandem, in a Scanning Mobility Particle Sizer spectrometer configuration (SMPS 3936, TSI), to measure particle number size distributions in the sub-micrometric range.

2.3 Methodology

Experiments were designed to: i) measure both cluster ion and particle number concentrations in the indoor microenvironment; and ii) evaluate the charging characteristics of particles emitted by different cooking activities through total positively-charged particle measurements, and charge specific size distribution measurements.

2.3.1 Cluster ion and particle concentration measurements

Cluster ion concentrations and total particle number concentrations were measured during three different cooking processes: a) gas stove at full power (no food, no pan); b) pan heating with gas stove at full power (no food); and c) grilling of bacon (100 g) with the gas stove operating at full power. Each cooking/combustion activity was conducted over a period of 10 min. The background particle number and cluster ion concentrations were monitored for 10 min before each cooking experiment. The measurements were continued during the cooking activity itself and over the following 30-40 min after cooking had ceased. The pan was cleaned before each experiment to remove any food residue due to previous tests. The experimental apparatus (a CPC and two AICs) were placed at a horizontal distance of 2 m from the stove. Positive and negative cluster ion concentrations were measured with a 1-min time resolution by two AICs, while total particle number concentration was measured by the CPC using an aerosol sampling air flow rate of 0.3 L min^{-1} and recorded every 5 s. Particle emission rates were also evaluated for the three tests performed on the basis of the procedure proposed by He et al. (2004) and used in our previous paper (Buonanno et al., 2009) and the abovementioned AER value. Emission rate data reported in the result section represent the mean value of three tests.

2.3.2 Total positively-charged particle measurements

Total positively-charged particle concentration (also known as composite size distributions of positively charged particles; Maricq (2006)) in the air during gas combustion, bacon grilling (100 g), and eggplant grilling (100 g) events was measured and compared to the overall particle size distribution to determine the pre-existing particle charge distribution of such aerosols. Both fat rich (bacon) and vegetable (eggplant) foods were considered as they are recognized emitting particles with different amount of volatile and semivolatile compounds (Buonanno et al., 2011b).

The cooking procedure was identical to that described in section 2.3.1. Indoor aerosol concentration was monitored throughout the cooking activities by two SMPS 3936 spectrometers. In particular, overall particle size distribution was measured by the SMPS made up of the Electrostatic Classifier with the radioactive aerosol neutralizer (3077A), while the positively-charged particle distribution was measured by the second SMPS used without a neutralizer, in order to sample only the particles that were already positively charged during the combustion process (Maricq, 2006). Particle number distributions were measured by the SMPS with a scan time of 135 s (including a retrace time of 15 s). During the gas combustion and eggplant grilling tests the SMPS aerosol and sheath flow rates were set to 1.5 and 15.0 L min^{-1}, respectively, yielding particle number distributions in the size range 6-220 nm. In all other experiments, SMPS aerosol and sheath flow rates were set to 0.3 and 3.0 L min^{-1}, respectively, during bacon grilling tests then obtaining a particle number distributions in the size range 14-700 nm. Diffusion loss correction was applied through the Aerosol

Instrument Manager Software TSI Inc. for both the SMPS spectrometers, whereas multiple charge correction (Hoppel, 1978) was applied only for the SMPS with neutralizer. Particle number concentrations were calculated from particle distribution data and measurement results represent the mean value of three tests.

Positively-charged particle distribution (i.e. the unknown pre-existing positive charge distribution) was obtained comparing the charge-neutral distribution measured through the SMPS with neutralizer to the distribution measured through the SMPS without neutralizer as reported by Buckley (2008) and Maricq (2006). To this purpose, the latter particle size distribution was uncorrected for the Gunn-Wiedensohler charging efficiency automatically applied by the Aerosol Instrument Manager Software TSI Inc. (Gunn, 1956; Wiedensohler, 1988b, a). Moreover, this uncorrected distribution was manually corrected for multiple charges applying the method reported in Hoppel (1978): a room temperature bipolar equilibrium particle charge distribution was considered applying the manual multiple charge correction for indoor aged aerosol and gas combustion, since aerosol emitted by flame suddenly reaches the room temperature Boltzmann distribution when they leave the flame itself (Maricq, 2008). Otherwise, bacon and eggplant grilling distributions were corrected applying a multiple charge correction derived for such specific aerosol as described in 2.3.3 and whose results are summarized in 3.3.

2.3.3 Particle charge distribution measurements

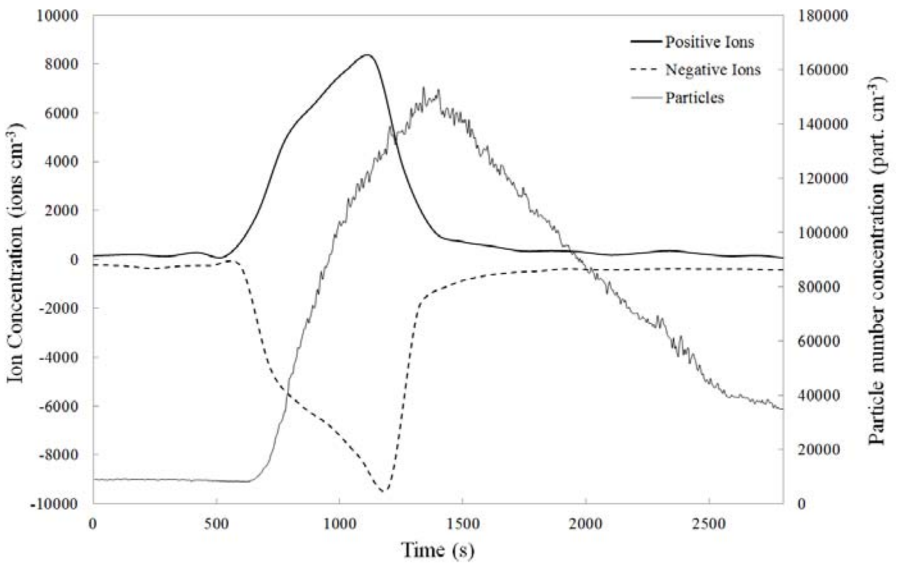
Charge specific size distribution was determined by the TDMA system, which has been described and employed in previous works (Buckley, 2008; Kim, 2005; Lee et al., 2012; Maricq, 2006). The TDMA system consists of an EC operated without a neutralizer and a SMPS spectrometer. The sampled particles pass through the first classifier without undergoing any neutralization. Therefore, they enter the DMA region carrying their own charge. A fixed voltage is applied through the first EC so that particles having the corresponding mobility diameter are selected. In this way, particles having a larger physical diameter but carrying a higher number of positive charges will be classified with the same mobility diameter (Buckley, 2008; Kim, 2005). The selected particles were passed to the SMPS spectrometer, where a typical 135-s dynamic scan was performed in the range 14-800 nm, thereby providing a particle distribution spectrum with several narrow peaks. The main peak is characteristic of single positively-charged particles of the original selected size, whereas the minor peaks represent larger particles having the same mobility diameter selected through the EC but carrying multiple positive charges (Buckley, 2008; Kim, 2005; Wiedensohler, 1988a).

Charge specific size distributions for bacon and eggplant grilling were evaluated choosing four particle diameters: 30, 50, 80 and 100 nm. Five 135-s SMPS size scans were obtained for each diameter. Sampling was conducted only during cooking activities, with decay concentrations not considered in these experiments, in order to avoid any ageing aerosol processes eventually leading to artifacts in charge distribution evaluation. The average percentage of positively charged particle concentration was derived.

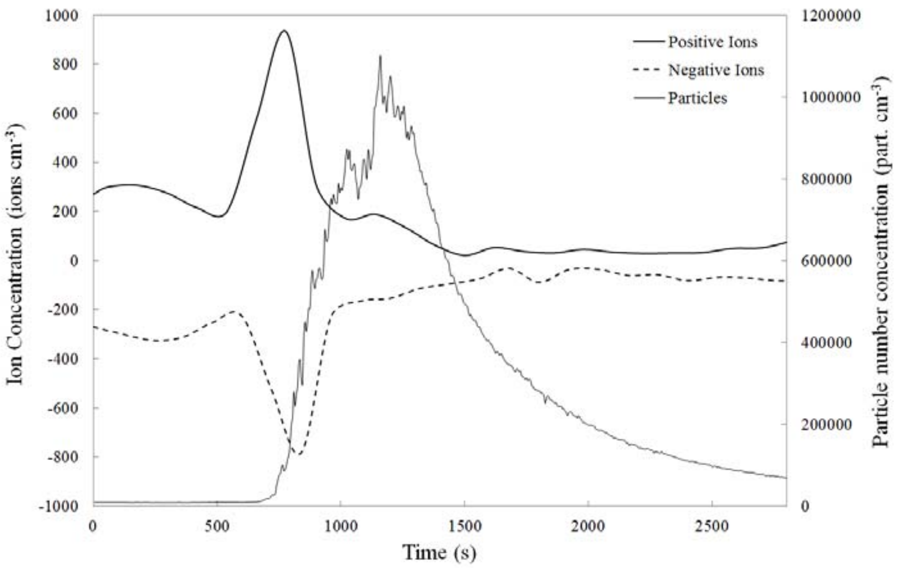
3. Results and discussions

3.1 Cluster ion and particle number concentration

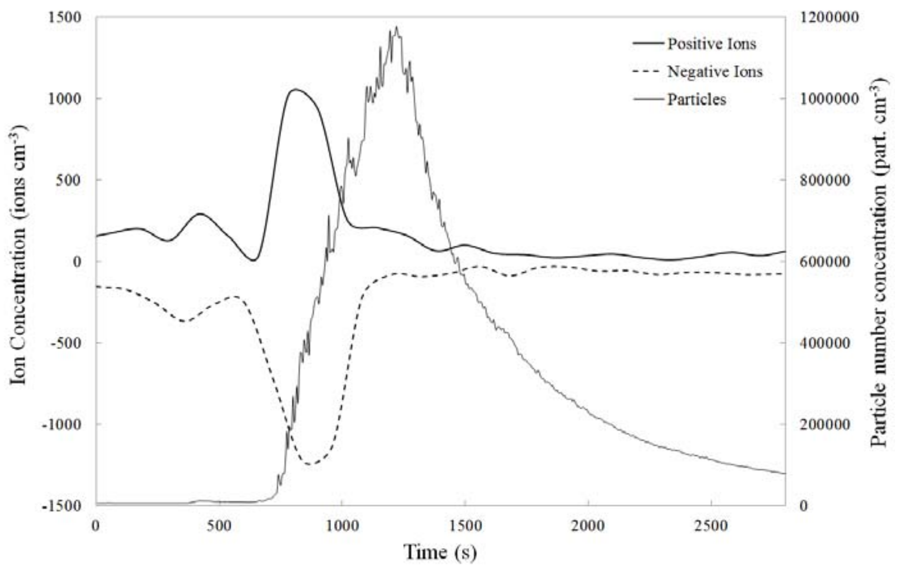
Time series of average cluster ion and particle number concentrations measured during the three cooking events are presented in Figure 1, while peak concentration data are summarized in Table 1. Total particle number concentration peaks of 1.54×10^{5}, 1.10×10^{6} and 1.18×10^{6} part. cm^{-3} were measured for gas combustion, pan heating and bacon grilling events, respectively. The standard deviations of particle number concentration peaks were found to be smaller than 20%, demonstrating the good repeatability of the experiments. The corresponding average particle emission rates were 1.73×10^{12}, 1.16×10^{13}, and 1.19×10^{13} part. min^{-1} for gas combustion, pan heating and bacon grilling events, respectively. The highest emission rates were measured for pan heating and bacon grilling events, and they were about one order of magnitude higher than during gas combustion alone (Buonanno et al., 2009; He et al., 2004; Wallace et al., 2008).


Positive Ions
Negative Ions
Particles
Ion Concentration (ions cm-3)
Particle number concentration (part. cm-3)
Time (s)

a)


Positive Ions
Negative Ions
Particles
Ion Concentration (ions cm-3)
Particle number concentration (part. cm-3)
Time (s)

b)


Positive Ions
Negative Ions
Particles
Ion Concentration (ions cm-3)
Particle number concentration (part. cm-3)
Time (s)

c)

Figure 1 – Temporal evolutions of cluster ion and particle number concentrations measured during the three tests: a) gas alone, b) pan heating, and c) 100 g bacon grilling. Trends represent the average of three measurements.

Table 1 – Particle number and cluster ion concentrations and emission rates measured during the three different cooking processes. Data represent the average of three tests.

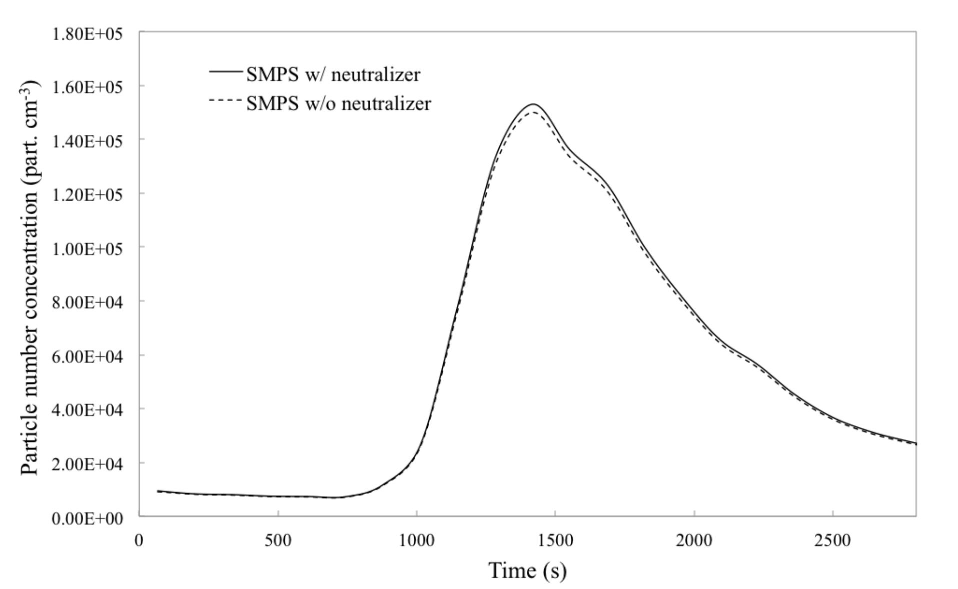
	Particle concentration peak (part. cm^{-3})	Particle emission rate (part. min^{-1})	Negative cluster ion concentration peak ($^{(-)}$ions cm^{-3})	Positive cluster ion concentration peak ($^{(+)}$ions cm^{-3})
Gas combustion	$1.54 \pm 0.21 \times 10^5$	1.73×10^{12}	$9.29 \pm 2.87 \times 10^3$	$8.18 \pm 1.32 \times 10^3$
Pan heating	$1.10 \pm 0.21 \times 10^6$	1.16×10^{13}	$7.8 \pm 1.9 \times 10^2$	$9.3 \pm 2.6 \times 10^2$
Bacon grilling	$1.18 \pm 0.06 \times 10^6$	1.19×10^{13}	$1.23 \pm 0.25 \times 10^3$	$1.02 \pm 0.18 \times 10^3$

Symmetrical positive and negative cluster ion concentrations were measured in all of the experiments performed, this being typical of combustion sources (Jayaratne et al., 2010; Maricq, 2005). The peak concentrations strongly varied from source to source: the positive/negative cluster ion concentration peaks measured for gas combustion, pan heating and bacon grilling events were $8.18 \times 10^3/9.29 \times 10^3$ ions cm^{-3}, $9.3 \times 10^2/7.8 \times 10^2$ ions cm^{-3}, and $1.02 \times 10^3/1.23 \times 10^3$ ions cm^{-3}, respectively. These values are higher than those measured both indoors and outdoors, in the absence of natural and anthropogenic sources (Hirsikko, 2011; Jayaratne et al., 2008; Jayaratne et al., 2010). In particular, gas combustion result in indoor cluster ion concentrations eight- to ten-fold larger than the ones involving grilling activities (pan heating and bacon grilling). One possible reason for such a marked variation of indoor cluster ion concentrations could be attributed to the difference in indoor particle concentrations generated by these three types of activities. The higher the number of particles emitted and measured, the larger the probability for cluster ions to attach to the particle surface, therefore, more particles will be charged and the number of cluster ions will decrease (Hoppel, 1985; López-Yglesias and Flagan, 2013). This is also confirmed by the trends reported in Figure 1, where cluster ion concentrations peaks are closer to particle concentration peaks during gas combustion when compared to the other two events. In particular, during pan heating and bacon grilling, the strong emission of particles leads to a sharp decrease in cluster ion concentrations. The time interval between cluster ion and particle number concentration peaks during activities involving grilling were about twice as long as during gas combustion (3 min vs. 6 min).

It is to be expected that indoor ion concentrations could not only be influenced by the emission rate of the source, but also by the size distribution of the particles emitted. In fact, as reported in the following sections (and discussed in previous work, Buonanno et al. (2009)), gas combustion particle distribution exhibits a size mode much lower than those measured during grilling activities. This leads to a possible lower surface area available for cluster ions to attach on to particles. This is in agreement with the main mechanism involved in aerosol charging during combustion processes, which has been identified as ion-particle collision (Kittelson, 1986; Maricq, 2006).

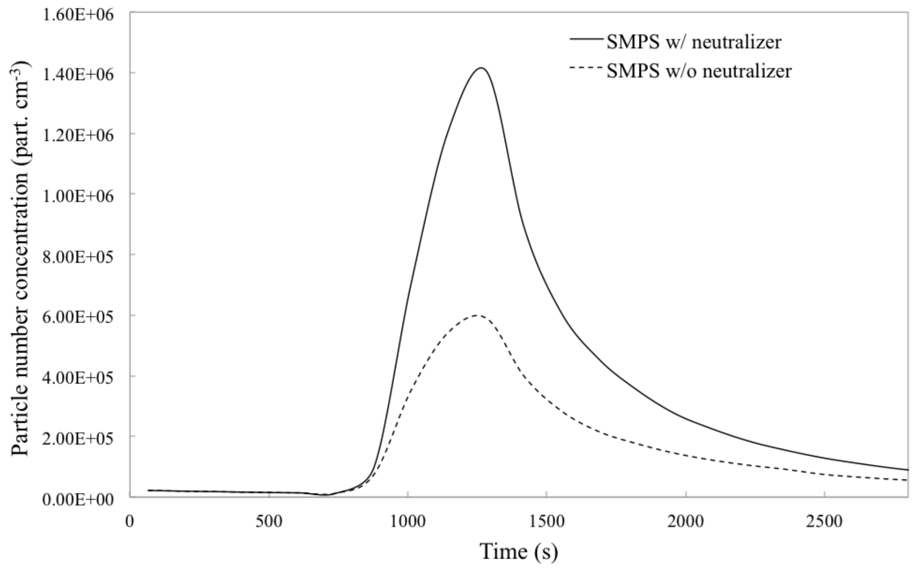
3.2 Total positively-charged particle distribution

In Figure 2, average trends of total particle number concentrations (measured using the SMPS 3936 with a neutralizer) were compared to the total particle number concentrations measured through the SMPS without neutralizer. The latter still includes the automatic charging efficiency correction as applied by the AIM software. This concentration was then unadjusted for charging efficiency and manually corrected for multiple charges, as reported in the section 2.3.2, in order to evaluate the percentage of positively charged particles emitted by each particle generation event. Figure 2 shows that simultaneous peaks in total particle number trends both with and without aerosol neutralization were measured for all the cooking events investigated. Total particle number concentration trends obtained from the SMPS were similar to the ones obtained from the CPC reported in the section 3.1. The maximum total particle number concentrations measured for the gas combustion, bacon grilling and eggplant grilling events were $1.53 \pm 0.11 \times 10^5$ and $1.41 \pm 0.18 \times 10^6$ part. cm^{-3}, and $1.13 \pm 0.15 \times 10^6$ respectively. Total particle number concentrations measured by the SMPS without neutralizer present a different behavior depending on the cooking event under investigation. In particular, it is equal to the one measured through the SMPS with neutralizer for the gas combustion event whereas is much lower for the two grilling events. As hereinafter reported, this is due to the different charging distribution of particles emitted by gas combustion compared to the ones generated by bacon and eggplant grilling events. The latter are characterized by a particle charge distribution giving particle charges that are lower than that predicted by the Boltzmann distribution, whereas gas combustion produces aerosol consistent with the Boltzmann charging distribution (Buckley, 2008; Maricq, 2005, 2006, 2008).



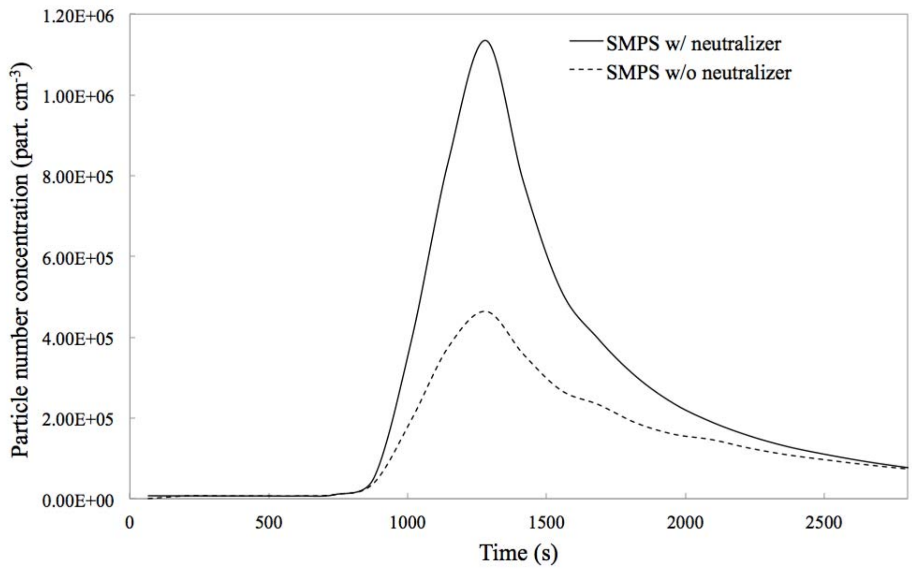
SMPS w/ neutralizer
SMPS w/o neutralizer
Particle number concentration (part. cm-3)
1.80E+05
1.60E+05
1.40E+05
1.20E+05
1.00E+05
8.00E+04
6.00E+04
4.00E+04
2.00E+04
0.00E+00
0
500
1000
1500
2000
2500
Time (s)

a)



SMPS w/ neutralizer
SMPS w/o neutralizer
Particle number concentration (part. cm-3)
1.60E+06
1.40E+06
1.20E+06
1.00E+06
8.00E+05
6.00E+05
4.00E+05
2.00E+05
0.00E+00
0
500
1000
1500
2000
2500
Time (s)

b)

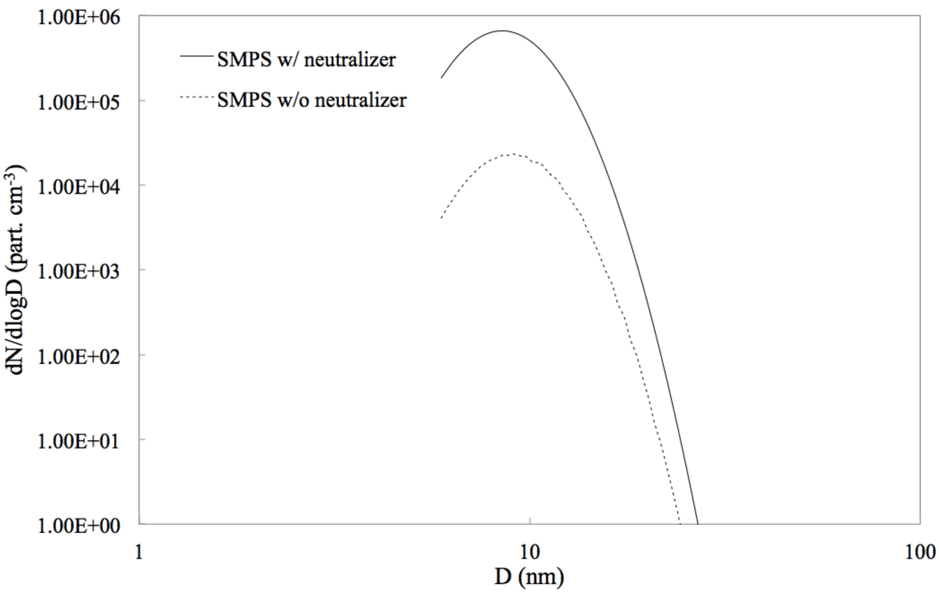


SMPS w/ neutralizer
SMPS w/o neutralizer
Particle number concentration (part. cm-3)
1.20E+06
1.00E+06
8.00E+05
6.00E+05
4.00E+05
2.00E+05
0.00E+00
0
500
1000
1500
2000
2500
Time (s)

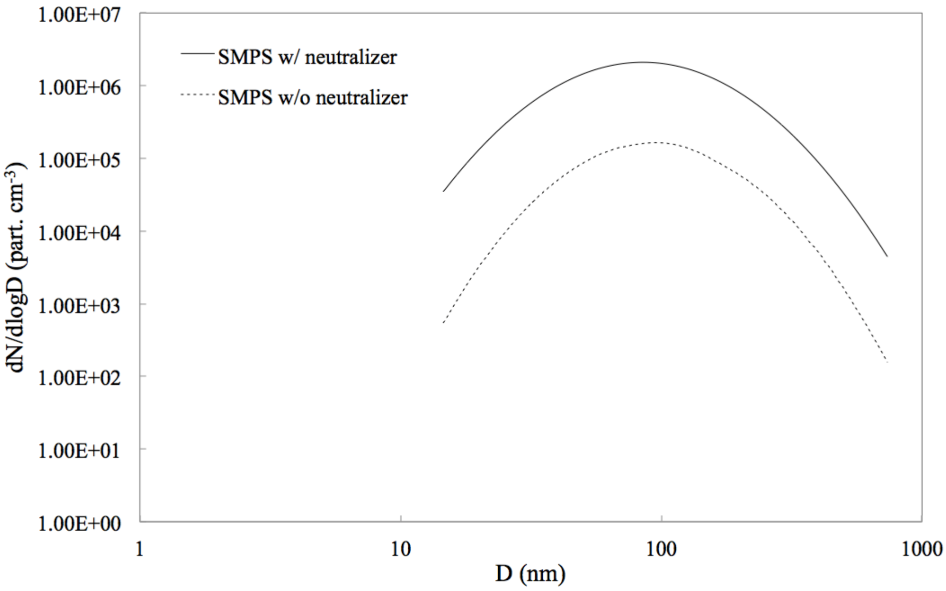
c)

Figure 2 – Time series of the total particle number concentration measured by the SMPS with (w/ neutralizer) and without (w/o neutralizer) neutralizer for gas alone (a), 100 g bacon grilling (b), and 100 g eggplant grilling (c) tests. Data measured by the SMPS without neutralizer include the charging efficiency function automatically applied by the AIM software (Gunn, 1956; Wiedensohler, 1988b, a).

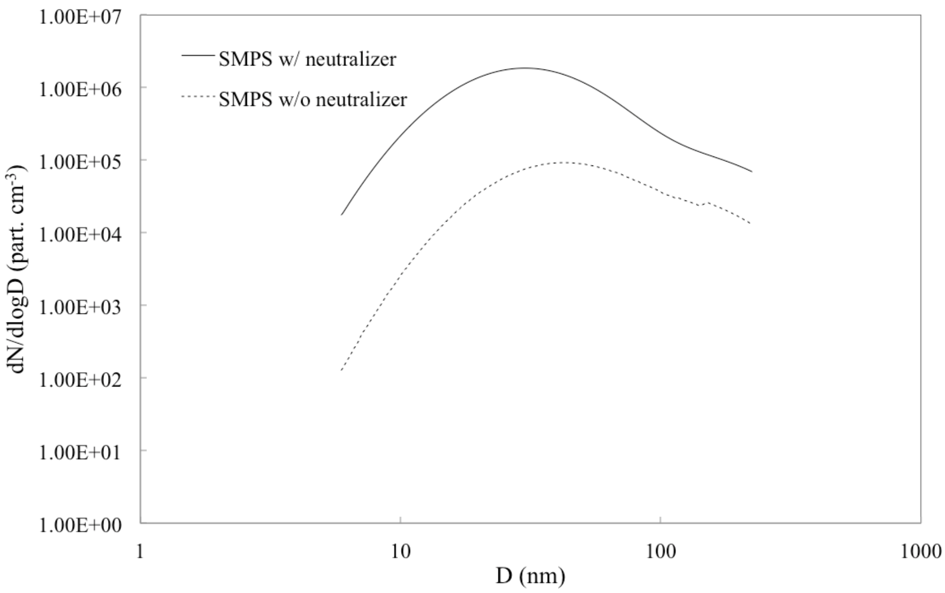
In Figure 3 the particle number distributions measured using SMPS, both with and without neutralizer, are reported for the cooking events under investigation. They represent the average distributions (of three tests) occurring at the same time as the total particle number concentration peaks (Figure 2). In particular, maximum distributions for gas combustion, bacon and eggplant grilling events (Figure 3a-c) are reported. Particle distributions when no sources were in operation (before bacon grilling) are also reported in Figure 3d. The distributions obtained through the SMPS with neutralizer were corrected for diffusion and multiple charge, whereas the distributions obtained from SMPS without neutralizer, corrected for diffusion losses, are unadjusted for the charging efficiency automatically applied by the AIM Software and manually corrected for multiple charges as reported in the section 2.3.2. Figure 3 shows that particle number distributions strongly depend on the type of cooking activity. For example, gas combustion events exhibited a size mode at 8 nm (which is in agreement with the findings of Wallace et al. (2008)) and 99.9% of the corresponding total particle number is in the range below 20 nm. On the contrary, larger particles are emitted during grilling events, in particular, bacon and eggplant grilling tests present average particle size modes of 85 and 30 nm, respectively. Average modes of positively-charged particle number distributions were found equal to 9, 88, and 34 nm gas combustion, bacon grilling, and eggplant grilling experiments, respectively.



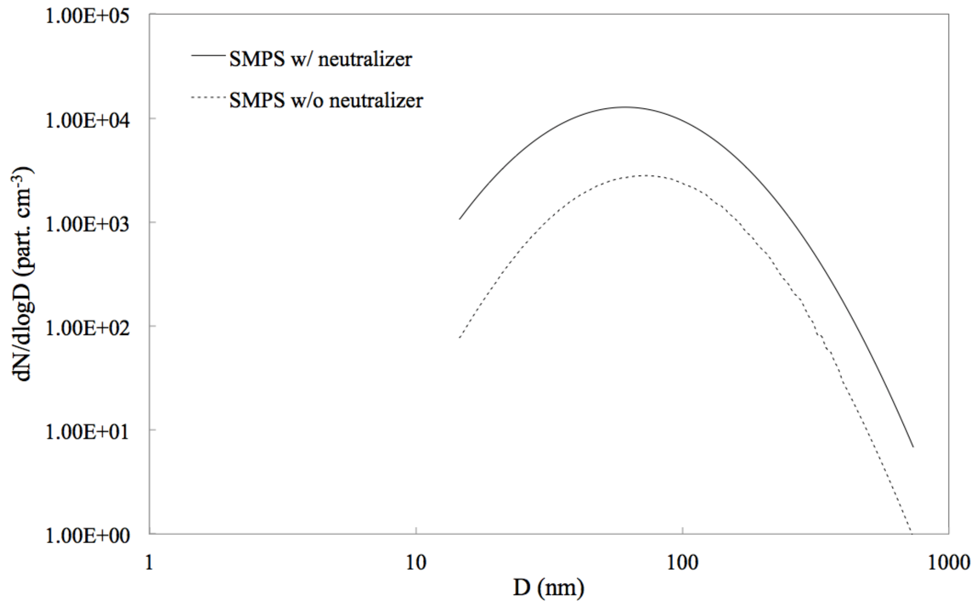
a)


SMPS w/ neutralizer
SMPS w/o neutralizer
dN/dlogD (part. cm^{-3})
1.00E+07
1.00E+06
1.00E+05
1.00E+04
1.00E+03
1.00E+02
1.00E+01
1.00E+00
1
10
100
1000
D (nm)

b)


SMPS w/ neutralizer
SMPS w/o neutralizer
dN/dlogD (part. cm^{-3})
1.00E+07
1.00E+06
1.00E+05
1.00E+04
1.00E+03
1.00E+02
1.00E+01
1.00E+00
1
10
100
1000
D (nm)

c)

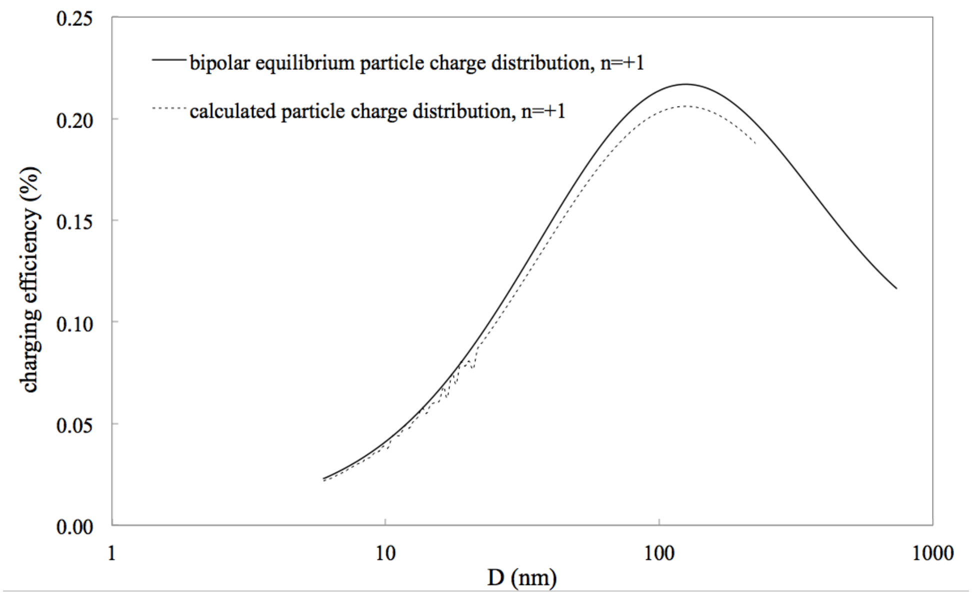


d)

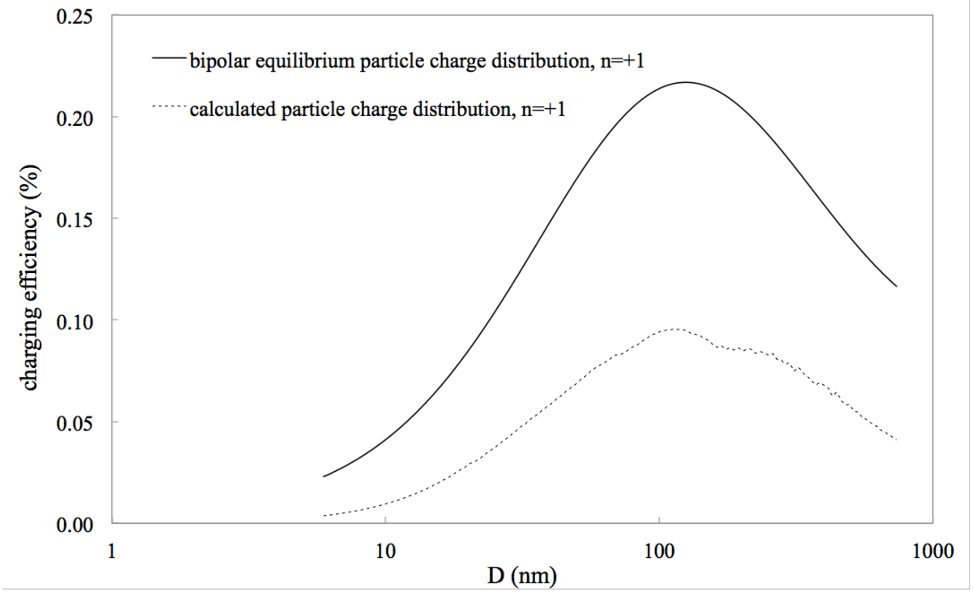
Figure 3 – Comparison between particle number size distribution measured by means of the SMPS with (w/ neutralizer) and without (w/o neutralizer) neutralizer for gas alone (a), 100 g bacon grilling test (b), 100 g eggplant grilling tests (c), and indoor ambient particles before cooking (d). Particle number distributions obtained using the SMPS 3936 with neutralizer are corrected for diffusion and multiple charges, whereas the ones reported w/o neutralizer represent the distributions corrected for diffusion losses, unadjusted for Gunn-Wiedensohler charging efficiency automatically applied by the AIM Software and then manually corrected for multiple charges (Hoppel, 1978).

In Figure 4 the singly charged positive particle distribution measured for gas alone, bacon grilling, eggplant grilling tests, and indoor ambient particles before cooking (obtained on the basis of the particle distributions reported in Figure 3, (Buckley, 2008)) are compared to the singly charged (n=+1) bipolar equilibrium particle charge distribution in air at room temperature (Gunn, 1956; Wiedensohler, 1988a). Such data clearly show that indoor particles, when no sources are in operation, result charged with a distribution quite similar to the bipolar equilibrium particle charge distribution per se. This is the reason why the particle concentration trends measured with and without neutralizer are identical before particle generation processes from cooking (Figure 2). During gas combustion the particle charge distribution is still similar to the Boltzmann distribution as also shown by the similar concentration trends in Figure 2a. In fact, combustion-generated particles are recognized to be neutrally charged according the Boltzmann charge distribution (Buckley, 2008; Maricq, 2005, 2006, 2008). On the contrary, singly charged positive particle distribution characteristics of bacon and eggplant grilling events, here shown on the maximum particle concentration, are much lower than the bipolar equilibrium particle charge distribution in air (Gunn, 1956; Wiedensohler, 1988a). This explains the gap between particle concentration trends measured by the SMPS with and without neutralizer for grilling events in Figure 2b and Figure 2c: in fact, the maximum difference occurred at the particle concentration peak, i.e. when the influence of freshly-generated particles is highest. The gap decreases during the particle concentration decay due to the particle ageing process leading to the same particle concentration level (and charge) measured before cooking. The difference in aerosol charging of the aerosol under investigation are better highlighted and quantified in Table 2, where the fractions of singly charged positive particles occurring at 10, 50, 100, and 200 nm mobility diameter are summarized for all tests performed and compared to the bipolar equilibrium particle charge distribution in air (Gunn, 1956; Wiedensohler, 1988a). The charge state lower than the expected Boltzmann distribution, which is typical of soot particles emitted during combustion processes, could be addressed to the volatile and semivolatile compounds condensing onto the soot particles (Buonanno et al., 2011b), then suddenly growing without changing their charge and, thereby lowering the charge fraction with respect to the Boltzmann distribution.

The authors point out that similar experimental analyses should be performed to investigate the behavior of negatively charged particles using a positive high-voltage power supply. Anyway, since i) the sources under investigation present a symmetrical emission of positive and negative ions and ii) the mobility of positive and negative ions is quite similar, the lower aerosol charge level (here shown when cooking activities are in operation) cannot be addressed to a shift to negatively charged particles.



a)


0.25
0.20
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charging efficiency (%)
bipolar equilibrium particle charge distribution, n=+1
calculated particle charge distribution, n=+1
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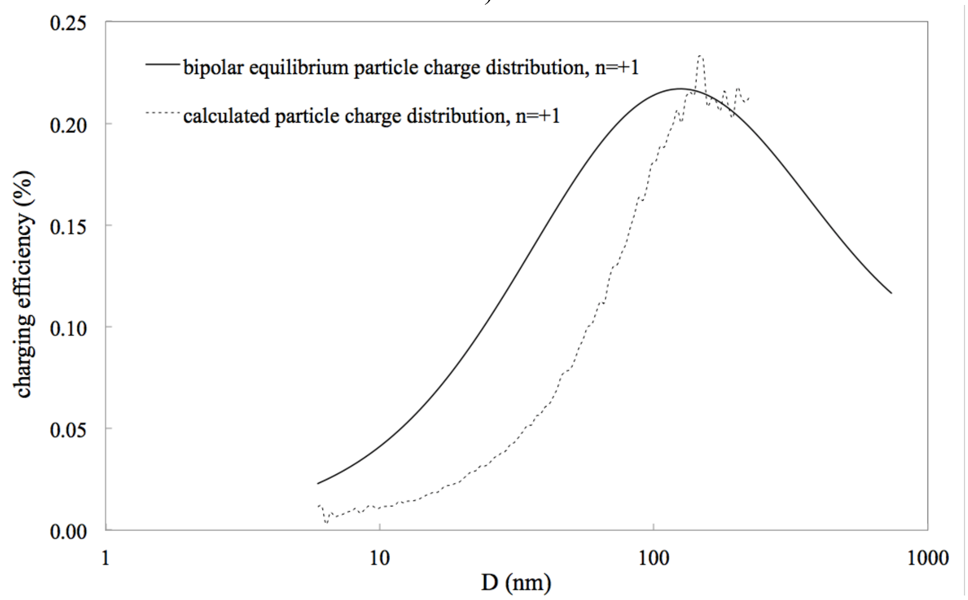
10
100
1000
D (nm)

b)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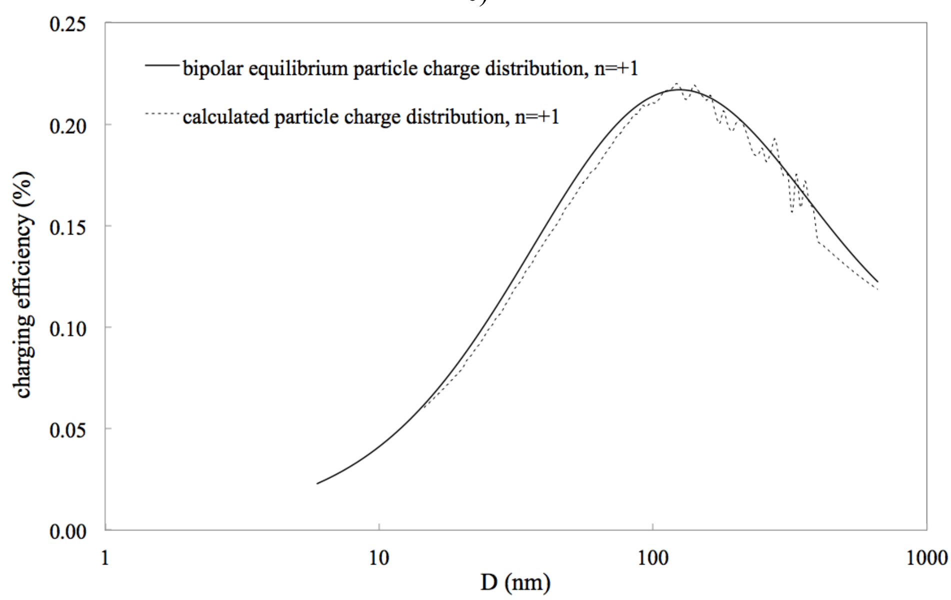
D (nm)

c)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bipolar equilibrium particle charge distribution, n=+1
calculated particle charge distribution, n=+1
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D (nm)

d)

Figure 4 – Comparison between singly charged positive particle distribution measured for gas alone (a), 100 g bacon grilling test (b), 100 g eggplant grilling tests (c), and indoor ambient particles before cooking (d) with bipolar equilibrium particle charge distribution in air (Gunn, 1956; Wiedensohler, 1988a).

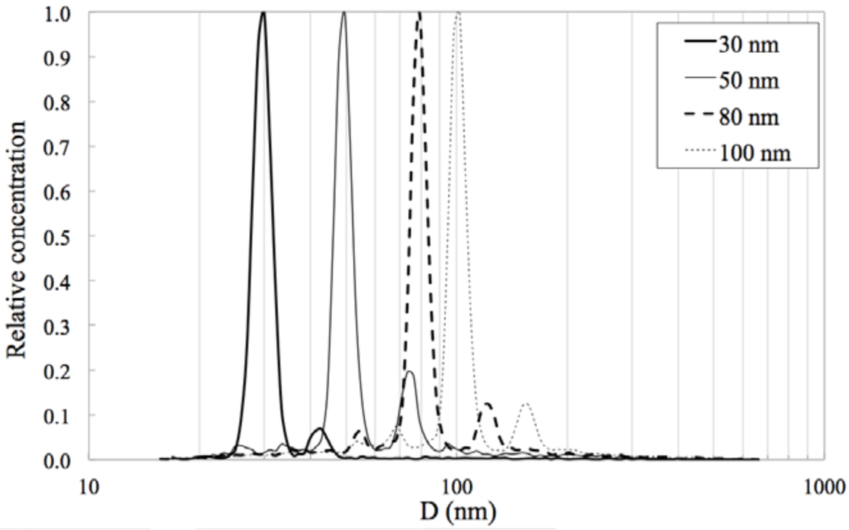
Table 2 – Fraction of singly charged positive particles measured for gas alone, bacon grilling test, eggplant grilling tests, and indoor ambient particles before cooking.. Comparison with bipolar equilibrium particle charge distribution in air (Gunn, 1956; Wiedensohler, 1988a)

D (nm)	equilibrium particle charge distribution (n=+1)	gas combustion (peak)	bacon grilling	eggplant grilling	before cooking
10	4.2%	3.8%	1.0%	1.2%	4.3%
50	16.9%	16.0%	6.9%	7.9%	16.1%
100	21.4%	21.4%	9.5%	18.1%	21.0%
200	20.4%	20.4%	8.5%	21.8%	20.1%

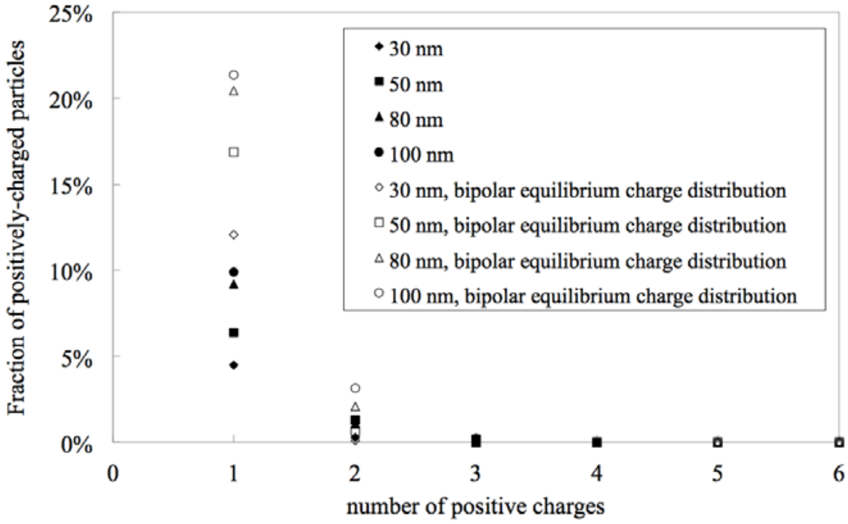
Finally, the percentage of total positively-charged particles, with respect to the total number of particles, has also been evaluated comparing the total particle number concentrations obtained from the distributions with and without neutralizer on the particle concentration peak discussed in Figure 3. In particular, the fraction of total positively-charged particles during gas combustion, bacon grilling, and eggplant grilling events was measured equal to 4.0%, 7.9% and 5.6%, respectively. On the contrary, the fraction of total positively-charged particles of indoor aged aerosol (before cooking event) was 17.4%. Similar laboratory-based tests performed by Kittelson (1986) and Maricq (2006) on diesel-emission particles resulted in approximately 30% of both positively- and negatively-charged particles each and about 40% of neutral particles. Therefore, particles emitted during grilling are, on average, much less charged than freshly-emitted diesel particles as well as those measured downwind of freeways with different type of traffic (Lee et al., 2012).

3.3 Particle charge distribution results

In order to evaluate the number of charges carried by particles of different sizes, charge specific size distribution for bacon and eggplant grilling was also determined by the TDMA system, as described in section 2.3.3. Similar results were obtained for bacon and eggplant grilling events. As example are here discussed the bacon grilling results. In Figure 5a, the average charge specific size distributions for 30, 50, 80 and 100 nm particles generated during bacon grilling experiments are shown in terms of relative concentrations, normalized to the peak particle number concentration. Peak characteristics of singly and multiply charged particles are shown. In particular, the four main peaks coincide with the selected diameter, as they represent 30, 50, 80, and 100 nm particles carrying one positive charge (+1). Smaller peaks were measured for particles carrying multiple charges (+2, +3, +4): they are characteristics of larger particles (e.g. 43, 74, 113, and 146 nm for 30, 50, 80, and 100 nm selected particles carrying two positive charges, respectively). In Table 3 and Figure 5b, the fractions of total particle concentration carrying +1, +2, +3 and +4 elementary charges are shown as a function of electrical mobility particle diameter. This size specific data shows that almost all of the positively-charged particles are singly charged: the significant difference with respect to equilibrium charge bipolar distribution was also highlighted. Charge distribution data obtained for bacon and eggplant grilling events were fitted and used in the manual multiple charge correction discussed in the sections 2.3.2 and 3.2.



a)



b)

Figure 5 – Charged-particle fraction distributions measured during bacon grilling experiments: a) normalized size distributions of positively-charged particles of different electrical mobility diameter (30, 50, 80, and 100 nm); b) comparison with bipolar equilibrium particle charge distribution in air (Gunn, 1956; Wiedensohler, 1988a) in terms of charge specific distribution.

Table 3 – Charge distribution of particles emitted during bacon grilling cooking activity: fraction of particle singly and multiply charged.

	Fraction of positive charges			
D (nm)	**+1**	**+2**	**+3**	**+4**
30	4.5%	0.3%	0.0%	0.0%
50	6.4%	1.3%	0.2%	0.1%
80	9.0%	1.1%	0.2%	0.1%
100	9.7%	1.2%	0.2%	0.1%

Conclusion

The aim of this paper was to evaluate the charging state of cooking-generated particles. This is because, even if the aerosol dose received by humans in indoor environments during cooking activities has been previously estimated, the effect of charges on these particles has not been considered. To this purpose an experimental analysis was designed to evaluate both cluster ion and particle number concentrations in the indoor microenvironments as well as charging characteristics of particles emitted by different cooking activities through total positively-charged particle measurements, and charge specific size distribution measurements.

Particle number concentration measurements allowed to evaluate average particle emission rates equal to 1.73×10^{12}, 1.16×10^{13}, and 1.19×10^{13} part. min^{-1} for gas combustion, pan heating and bacon grilling events, respectively. Simultaneous ion concentration measurements revealed symmetrical positive and negative cluster ion concentration trends as typical of combustion sources: highest concentrations were measured during gas combustion possibly due to the lower particle surface area available for cluster ions attaching and removal.

Simultaneous measurements of particle size distributions using SMPS spectrometers with and without neutralizer were performed to evaluate the pre-existing particle charge state of aerosol produced during gas combustion, bacon grilling and eggplant grilling events. The charge distribution of particle emitted by gas combustion was recognized similar to the bipolar equilibrium charge distribution, whereas aerosols produced during grilling events present charge state much lower than the one predicted by the bipolar equilibrium charge distribution. This could be due to the volatile and semivolatile compound condensing onto particles emitted then lowering their charge fraction. In particular, the fraction of total positively-charged particles during gas combustion, bacon grilling, and eggplant grilling events was measured equal to 4.0%, 7.9% and 5.6%, respectively. Thus, the charge state of cooking generated is lower than diesel-generated particles.

The authors point out that the present work contributes to involve the indoor sources/micro-environments in particle charging characterization research, so far essentially focused on outdoor sources and environments. The data here discussed, along with previously deepened physical-chemical properties, contribute to improve the knowledge on cooking-generated particles. The present results could be applied, for example, in the evaluation of particle doses experienced during cooking activities then representing an advance in our understanding of the associated dose-response relationships. Moreover, particle charge characterization is a key aspect in aerosol measurement field since several particle monitors can be affected by the pre-existing aerosol charge state.

Future researches will involve the evaluation of influential parameters on cooking-generated particle charge measurement, to this purpose different cooking methods (grilling, frying), type of heat source (gas or electric), kind of oil used to fry will be considered in order to exhaustively improve our knowledge of cooking-generated aerosol charging.

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