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Charged particles and cluster ions produced during cooking activities

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HIGHLIGHTS

- Electric charge of particles generated during cooking activities was measured.
- Positive and negative cluster ion concentration trends were symmetrical.
- Total fraction of positively-charged particles was lower than 10%.
- Gas combustion generates aerosols with a Boltzmann charge distribution.
- Cooking activities emit aerosols with charge state lower than the Boltzmann one.

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ABSTRACT

Previous studies showed that a significant number of the particles present in indoor air are generated by cooking activities, and measured particle concentrations and exposures have been used to estimate the related human dose. The dose evaluation can be affected by the particle charge level which is usually not considered in particle deposition models. To this purpose, in this paper we show, for the very 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, was 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, then lower than other typical outdoor combustion-generated particles.

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 (Ott, 1982) 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 (Berghmans et al., 2009; Buonanno et al., 2012a; 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.

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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 esthetic products Stabile et al., 2012), the main source of indoor UFPs is cooking activities. 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 of 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 ($>1 \times 10^5$ 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, 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. (2011), who performed particle number size 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.

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) are 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 et al., 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 of 200–2500 cm^{-3} (Hirsikko et al., 2011). In particular, concentrations of 300–400 cm^{-3} typically occur under stable atmospheric conditions, whereas natural (e.g. waterfalls, rainfalls Hirsikko et al., 2011; Laakso et al., 2007) and anthropogenic sources (e.g. powerlines, motor vehicles, trafficked roads Jayaratne et al., 2008, 2010, 2011; 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 et al., 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 shown. 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. A further aim of the present study is to improve the understanding of the deposition of indoor generated particles in the respiratory system. Since charged particles are more likely to be deposited in the human respiratory system than similarly sized uncharged particles, neglecting the particle charge characterization could result in wrong estimation of the overall dose received by humans in indoor environments.

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). In the present study both vegetable and fatty foods were tested in order to evaluate the possible effect of the food on the charge level of particles emitted during the cooking activities.