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Influential parameters on particle concentration and size distribution in the mainstream of e-cigarettes

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A B S T R A C T

Electronic cigarette-generated mainstream aerosols were characterized in terms of particle number concentrations and size distributions through a Condensation Particle Counter and a Fast Mobility Particle Sizer spectrometer, respectively. A thermodilution system was also used to properly sample and dilute the mainstream aerosol.

Different types of electronic cigarettes, liquid flavors, liquid nicotine contents, as well as different puffing times were tested. Conventional tobacco cigarettes were also investigated.

The total particle number concentration peak (for 2-s puff), averaged across the different electronic cigarette types and liquids, was measured equal to $4.39 \pm 0.42 \times 10^9$ part. cm^{-3}, then comparable to the conventional cigarette one ($3.14 \pm 0.61 \times 10^9$ part. cm^{-3}). Puffing times and nicotine contents were found to influence the particle concentration, whereas no significant differences were recognized in terms of flavors and types of cigarettes used.

Particle number distribution modes of the electronic cigarette-generated aerosol were in the 120–165 nm range, then similar to the conventional cigarette one.

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1. Introduction

Aerosol exposure is a major environmental health concern due to the particles' ability to penetrate deeply into the respiratory system and cell membranes (Unfried et al., 2007) and translocate from the airways into the blood circulation (Schins et al., 2004; Weichenthal, 2012). Particles are also able to deposit in secondary organ (Semmler et al., 2004), including brain tissue (Calderon-Garciduenas et al., 2004) and to carry condensed toxic compounds (Brown et al., 2001; Nygaard et al., 2004; Schmid et al., 2009). Concerning the human health, indoor air quality represents the most important issue since people spend most of their time indoors (Klepeis et al., 2001) where particle concentrations are typically high (Buonanno et al., 2013; EPA, 2004). A major indoor particle source is the environmental tobacco smoke, ETS (Nazaroff and Singer, 2004; Repace and Lowrey, 1980; WHO, 2005, 2013) which is a mixture of exhaled mainstream smoke, and sidestream smoke released from the smoldering tobacco products. Tobacco cigarettes contain around 4000 different chemicals, including toxins like arsenic and radioactive polonium-210 (Baker et al., 2004; Fowles and Dybing, 2003; IARC, 2004, 2012; Little et al., 1965; Wynder and Hoffmann, 1967). Moreover, in fresh unaged tobacco cigarette mainstream smokes were measured particle concentrations of about 4×10^9 part. cm^{-3}, with an arithmetic mean diameter of about 0.2 μm (Adam et al., 2009; Alderman and Ingebrethsen, 2011; Borgerding and Klus, 2005).

1.1. E-cigarettes: state-of-art

Nowadays, the use of electronic cigarettes (e-cigarettes) is becoming increasingly popular maybe because smokers consider it a healthier alternative to conventional smoking: anyway, comprehensive studies aimed to characterize the aerosol produced by these devices are still not available.

E-cigarettes are made up of three integrated parts contained in a stainless steel shell: a cartridge, an atomizer, and a battery. The cartridge is the liquid reservoir which also acts as a mouthpiece. When an e-cigarette smoker (named "vaper") inhales through the mouthpiece, an air flow sensor activates the atomizer, which heats up the liquid inside the cartridge producing a smoke-like vapor then orally inhaled (Riker et al., 2012). Liquid mixture consists of propylene glycol and/or vegetable glycerin, water, and flavors.

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Different nicotine concentration levels are commercialized: typically 0–6 mg mL^{-1} (low), 12–16 mg mL^{-1} (medium), and 18–24 mg mL^{-1} (high). Flavors can be both natural and artificial, moreover different flavor tastes are available such as tobacco, fruit, and herb. The e-cigarettes can be single-use (disposable, non-refillable) or reusable (refillable tank or not, welded tank atomizer or not), with either automatic or manual battery. Compared with conventional cigarettes, which last about fifteen puffs, e-cigarettes allow from 150 to 300 puffs (Wollscheid and Kremzner, 2009).

E-cigarette products are not adequately regulated so far (Gornall, 2012): the e-cigarette industry claims that the existing legislation (European Parliament and Council of the European Union, 2001a) and the EU rapid alert system, RAPEX, are adequate in their current form to regulate them as consumer products, while Tobacco Industry pushes to include e-cigarettes in the Tobacco Products Directive (TPD) (Roland Berger Strategy Consultants, 2013). The European Commission proposed to extend the TPD to nicotine containing product (NCP) regulation (it will be adopted in 2014), then including almost all e-cigarettes in the medicines regulation (European Parliament and Council of the European Union, 2001b). Thus, e-cigarettes will be required to obtain a marketing authorization from a health regulator.

Very few studies investigated health effects due to the e-cigarette use (Bullen et al., 2010; Dawkins et al., 2012; Flouris et al., 2013, 2012; Vansickel et al., 2010). A review of 16 studies (Cahn and Siegel, 2011) found e-cigarettes comparable in toxicity to nicotine replacement therapies (NRT) but less harmful than tobacco cigarettes. Nonetheless, there are still some questions about the safety of the chemicals in e-cigarette liquids, and the current lack of regulation means there is no way of verifying what actually is in them, especially with so many different brands suddenly entered the market and the variation in performance properties within brands detected by Williams and Talbot (2011).

The products of the e-cigarettes may contain ingredients that are known to be toxic to humans. As example, the propylene glycol, released in the vapor, is known to be responsible of upper airway irritations (Wieslander et al., 2001). Vardavas et al. (2012) reported adverse physiologic effects after short-term use of e-cigarette similar to some effects recognized in tobacco smoking. Gennimata et al. (2012) also showed that e-cigarette use causes potential harmful short-term effects on lung function.

A further issue to be controlled and regulated, is the real nicotine content in the liquid (Britton and McNeill, 2013; Grana, 2013). As example, the US Food and Drug Administration detected nicotine trace and others dangerous substances even in e-cigarettes classified as nicotine-free (FDA, 2009). This is not a trivial aspect, since nicotine can be toxic in high doses and can lead people to nicotine addiction then inducing them to use other tobacco products such as conventional cigarettes (Bell and Keane, 2012). No information on long-term health effects of e-cigarette use is still available.

1.2. Aims of the work

The present study is focused on the total particle number concentration and size distribution measurement of the mainstream aerosol generated by e-cigarettes. Data were analyzed and compared to those from a conventional tobacco cigarette. In order to propose an exhaustive characterization of the e-cigarette emission, different influence parameters such as type of the e-cigarette, flavor, nicotine content and puffing time were investigated. Measurements of particle number concentrations and size distributions were performed with a one-second-time resolution in order to identify the impact of the particles inhaled by e-cigarette vaper on human health and to put a new insight for assessing of respiratory dosimetry.

2. Materials and methods

2.1. Experimental campaign

Different types of e-cigarettes were tested: two rechargeable models (e-cigarettes A and B) and one disposable model (e-cigarette C). Their characteristics are summarized in Table 1. E-cigarettes were filled with different liquids in terms of flavor and nicotine content. Rechargeable models were cleaned with deionized water after each test in order to avoid possible liquid contamination. Two tobacco flavors, (Liquid 1) and (Liquid 4), an e-juice flavor, (Liquid 2), and a herb flavor (Liquid 3) were used. Three nicotine levels were tested: zero (0 mg mL^{-1}), medium (8–9 mg mL^{-1}), and high (12–18 mg mL^{-1}) concentrations. Details of the liquid characteristics are reported in Table 2. E-cigarettes were recently purchased and unused prior to testing. Batteries of the rechargeable models (e-cigarettes A and B) were fully charged before each experiment. Conventional tobacco cigarettes were also tested. In particular, cigarettes with a nicotine concentration equal to 0.8 mg per cigarette were considered (Table 2).

Measurements were performed in the European Accredited (EA) Laboratory of Industrial Measurements (LAMI) at the University of Cassino and Southern Lazio, Italy, where thermo-hygrometric conditions were continuously monitored, in order to guarantee temperature and relative humidity values equal to 20 ± 1 °C and 50 ± 10%, respectively.

2.2. Instrumentation and quality assurance

In order to measure total particle number concentrations and size distributions the following instruments were used:

- a TSI model 3775 Condensation Particle Counter (CPC) able to measure total particle number concentration down to 4 nm in diameter with a one-second time resolution;
- a TSI model 3091 Fast Mobility Particle Sizer (FMPS) spectrometer able to measure particles size distribution and total concentration in the range 5.6–560 nm through an electrical mobility technique involving multiple electrometers getting simultaneously signals from all particle sizes with a one-second-time resolution;
- a thermodilution system (two-step dilution) made up of a Rotating Disk Thermodiluter, RDTD (model 379020; Matter Engineering AG) (Hüglin et al., 1997) and a Thermal Conditioner Air Supply (model 379030; Matter Engineering AG) (Burtscher, 2005) allowing to ensure a proper sample conditioning during cigarette-generated particle number distribution and total concentration measurements. Temperature control is also allowed in the thermodilution section by a built-in heater with selectable temperatures;
- a TSI model 3080 Electrostatic Classifier (EC) able to select airborne particles of uniform size from a polydisperse source, resulting in a highly monodisperse aerosol. It is also used along with a CPC 3775 for particle size distribution measurements in Scanning Mobility Particle Sizer (SMPS) spectrometer configuration;
- a TSI model 4410 Flow meter to check flow rates in the tubing connecting the cigarette to the measuring devices.

The CPC was calibrated in the European Accredited Laboratory at the University of Cassino and Southern Lazio by comparison with a TSI 3068B Aerosol Electrometer using NaCl particles generated through a Submicrometer Aerosol Generator (TSI 3940) (Stabile et al., 2013).

2.3. Methodology description

The experimental campaigns were carried out during February–June 2013. Measurements of total particle number concentrations and particle size distributions were performed considering different types of cigarettes and liquids as hereinafter detailed.

Three puff profiles were considered for each test. Each puff profile was performed considering four consecutive puffs (puffing time of 2, 3, or 4 s) with a 30-s inter puff interval. The first puff was considered a "warm up" puff as it could lead to possible measurement errors when e-cigarettes were tested, as also reported in Ingebrethsen et al. (2012). The conventional tobacco cigarette were tested using the same procedure of e-cigarettes. The puffs for both electronic and conventional cigarettes were performed connecting the aerosol sampling line to the cigarette

Table 1
Characteristics of the e-cigarettes tested.

Sample	Delivery system
E-cigarette A	Tank system
E-cigarette B	Atomizer phantom
E-cigarette C	Cartom
Conventional tobacco cigarette	–

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