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Aerodynamic particle size distribution and dynamic properties in aerosols from electronic cigarettes

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ABSTRACT

Aerodynamic Particle Size Distribution (APSD) is an important factor governing if aerosols can be inhaled to different parts of a human's respiratory system. To understand the APSD of the aerosol produced from several brands of E-cigarettes, measurements using the Next Generation Pharmaceutical Impactor (NGI) were performed.

The NGI impactor was operated at two different temperature conditions, ambient room temperature condition (20–25 °C) and in a cold condition (4–8 °C). The relative humidity (RH) in the latter case, cooled impactor, was close to 100%, similar to the human's respiratory system. Obtained Mass Median Aerodynamic Diameters (MMAD) for the investigated electronic cigarettes were in the range 0.5–0.9 μm. The different NGI temperature conditions had a significant effect on the result and this is probably due to dynamic processes during the aerosol transport in the NGI pass ways.

For each NGI measurements performed the nicotine present in both the droplets and in the gaseous phase were measured. This is different from the previously published data using physical aerosol sizing techniques (such as Scanning Mobility Particle Sizer, SMPS), where obtained droplet sizes is a result of all chemical components in the aerosol, not only nicotine. In principle, it could be only excipients in the droplets and all nicotine in gaseous phase. Thus, impactor and a gas trap with nicotine specific quantification is a good complement for more thorough understanding of droplet size distributions of E-cigarette.

The APSD in vitro data was used in theoretical modeling using the Multiple-Path Particle Dosimetry Model (MPPD v 2.11), for estimation of the lung deposition. Using the model it was estimated that 75–90% of the nicotine droplets will be exhaled and 10–25% deposited in the respiratory system. However, when an aerosol is inhaled to a human's respiratory system it will be diluted and the evaporation of nicotine will increase. This will lead to overestimation of the fraction of exhaled nicotine using the MPPD model.

1. Introduction

During recent years there has been a tremendous evolvement of the sales and usage of electronic cigarettes and Adkinson et al. (2013) reports that in a survey 7.8% of smokers have tried E- cigarettes. This translates into several million E-cigarette users and the market is growing. Health advocates continue to study and debate E-cigarettes. The mechanism of nicotine delivery is not clear, the need for quality control and safety studies, efficacy as smoking cessation devices and advisability as smoking alternatives are areas

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debated. For a comprehensive review of different aspects of E-cigarettes (see Etter, 2013).

We believe that nicotine delivery to the user is a prerequisite for E-cigarettes to work as smoking cessation products and the aerosol droplet size may have a profound effect on this. Product attributes such as dose uniformity, APSD of the aerosol and distribution of nicotine between droplets and vapor phase are especially important. These aspects are starting to gain more attention but there is still fairly limited data available in the public domain. Investigation of purity and quality of commercially available e-liquids was presented in a paper by Etter, Zäther, and Svensson (2013). Dosing properties of electronic cigarettes have been investigated and presented in a paper by Goniewicz, Kuma, Gawron, Knysak, and Kosmider (2012).

Cigarettes have been studied regarding aerosol properties (Sahu, Tiwari, Bhangare, & Pandit, 2013) as well as the distribution of nicotine between the gas phase and bound within particles (Häger & Niessner, 1997; Lipowicza & Piad, 2004). For E-cigarettes, the distribution between molecular nicotine in the gas phase and droplet bound nicotine has only been discussed to a limited extent. We would expect that studies using the denuder technique are under way to further describe the properties of the aerosol delivered. Denuder measurements would most probably drive the nicotine equilibrium from the particle or droplet bound towards the gas phase during measurement. To give an answer on the initial fraction gas vs. particle/droplet bound, a complex model calculation would be required.

An NGI is a tool used for determining APSD of aerosols. It was specifically designed for pharmaceutical aerosols (Marpel, Roberts et al., 2004) and is recommended as one of the accepted apparatus for determining APSD in the European Pharmacopeia (EP) and United States Pharmacopeia (USP) for pharmaceutical aerosols from inhalers. Different size fractions are impacted in the different stages of the NGI and subsequently quantified by chemical assay. The cut off diameters of the stages were designed to be equidistant on a log normal scale to provide good characterization of the aerosol since the vast majority of aerosols show a log normal distribution (Marpel, Olson et al., 2004; Marpel et al., 2005). The NGI impactor is aerodynamically calibrated at 15, 30, 60, and at 100 L/min. If a measurement is performed between 30–100 L/min, the cut off values are calculated by using the cut off values at 60 L/min (see Pharmacopeias for details). The cut off values at the air flows used in this study are shown in Table 1. To analyze the aerosols produced by E-cigarettes modification of the conventional pharmacopoeia NGI method was required. The modification of the test is described in this study.

APSD and the dynamic properties of the aerosol cloud delivered from the electronic cigarettes is crucial to understand if electronic cigarettes are equivalent to conventional cigarettes for delivering nicotine to the respiratory system. This study, containing a novel analysis methodology, provides new knowledge on E-cigarette aerosols from several different brands.

2. Materials and methods

2.1. Material

All electronic cigarettes were purchased on internet webshops. In all cases two cartridges (ampoules) were tested but often with the same power pack (battery+heating element). The tested brands are shown in Table 2.

The NGI and its accessories were supplied by Copley Scientific, Nottingham, UK. The oxalic acid dihydrate, 99,6%, CAS 6153-56-6, was supplied from Swedhandling, Norrköping, Sweden. The filter used after the NGI outlet, Respirgard II, model 303, GE, was supplied by Medcore, Sweden.

2.2. Method description

The NGI collection cups where used without coating. When coating was used in the test, the APSD was biased due to gaseous nicotine being trapped, see Fig. 1. For the recovery and quantification of the droplet bound nicotine in the cups (after E-cigarette dosing), 10 and 20 ml of 0.05 M H_2SO_4 was added to the cups and NGI throat, respectively. Extraction of the deposited aerosol for about 5 min was sufficient.

The nicotine concentrations in the samples from NGI cups and throat were determined by UV–vis spectroscopy at 259 nm and standard solutions for the quantification were prepared by dissolving Nicotine hydrogen tartrate dihydrate in 0.5 M H_2SO_4.

The electronic cigarettes were encapsulated in a tube slightly larger than the electronic cigarette which facilitated free passage of air into the air intake nozzle of the mounted device. The tube was connected to an adjustable pneumatic dispenser via silicon tubing.

Table 1
Aerodynamically calibrated cut off values at different flow rates for an NGI Impactor.

Cut off values (μm)	15 L/min	30 L/min	60 L/min
Stage 1	14.1	11.72	8.1
Stage 2	8.61	6.40	4.5
Stage 3	5.39	3.99	2.8
Stage 4	3.30	2.30	1.7
Stage 5	2.08	1.36	0.9
Stage 6	1.36	0.83	0.6
Stage 7	0.98	0.54	0.3
MOC	Na	0.17	0.118