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Effect of variable power levels on the yield of total aerosol mass and formation of aldehydes in e-cigarette aerosols

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A B S T R A C T

The study objective was to determine the effect of variable power applied to the atomizer of refillable tank based e-cigarette (EC) devices. Five different devices were evaluated, each at four power levels. Aerosol yield results are reported for each set of 25 EC puffs, as mass/puff, and normalized for the power applied to the coil, in mass/watt. The range of aerosol produced on a per puff basis ranged from 1.5 to 28 mg, and, normalized for power applied to the coil, ranged from 0.27 to 1.1 mg/watt. Aerosol samples were also analyzed for the production of formaldehyde, acetaldehyde, and acrolein, as DNPH derivatives, at each power level. When reported on mass basis, three of the devices showed an increase in total aldehyde yield with increasing power applied to the coil, while two of the devices showed the opposite trend. The mass of formaldehyde, acetaldehyde, and acrolein produced per gram of total aerosol produced ranged from 0.01 to 7.3 mg/g, 0.006 to 5.8 mg/g, and <0.003 to 0.78 mg/g, respectively. These results were used to estimate daily exposure to formaldehyde, acetaldehyde, and acrolein from EC aerosols from specific devices, and were compared to estimated exposure from consumption of cigarettes, to occupational and workplace limits, and to previously reported results from other researchers.

1. Introduction

Electronic cigarettes (ECs) are becoming increasingly popular, with millions of users both in the US and in Europe (Pearson et al., 2012; Regan et al., 2013; Vardavas et al., 2014) and are often used as a replacement for combustible cigarette usage (Barbeau et al., 2013). Aldehydes including formaldehyde, acetaldehyde, and acrolein are known to form during heating of mixtures of glycerol (GLY) and propylene glycol (PG) (Flora et al., 2015; Lauterbach and Spencer, 2015; Ohta et al., 2011; Paschke et al., 2014; Uchiyama et al., 2013), the most common solvent formulation for EC liquids. These aldehydes are of concern since formaldehyde is classified by the International Agency for Research of Cancer (IARC) as a human carcinogen (Group 1) and acetaldehyde is classified as possibly carcinogenic to humans (Group 2B) (IARC, 2012). Acrolein causes irritation of the nasal cavity and damages the lining of the lung (USEPA, 2003). Glycerol has been shown to produce these three aldehydes due to thermal decomposition (pyrolysis) in temperature-dependent amounts (Paine et al., 2007), with small amounts of acrolein being formed in some ionic environments at 350 °C, and all three aldehydes being formed at 600 °C. The pathway for this pyrolysis is shown in Fig. 1, and it involves a free-radical dehydration of glycerol to form 3-hydroxyl-1-propen-1-ol, which tautomerizes to 3-hydroxylpropionaldehyde. This then loses another water in a free-radical mechanism to form acrolein. At higher temperatures 3-hydroxylpropionaldehyde can convert to formaldehyde and acetaldehyde, by way of a retro-aldol reaction, which easily cleaves the C2–C3 bond at >400 °C.

Because of these known decomposition products, one of the main concerns related to EC use is the inhalation of aldehydes contained in EC aerosol. Studies on relatively lower power, prefilled disposable devices have found that formaldehyde, acetaldehyde, and acrolein are produced at levels far lower in comparison to tobacco cigarette smoke (Bekki et al., 2014; Cheng, 2014; Goniewicz et al., 2014; Lauterbach and Spencer, 2015). However, recent studies on higher powered, refillable tank systems have found that these devices may produce levels of aldehydes exceeding the levels found in mainstream cigarette smoke (Jensen et al., 2015; Kosmider et al., 2014). To date, however, there has not been a systematic

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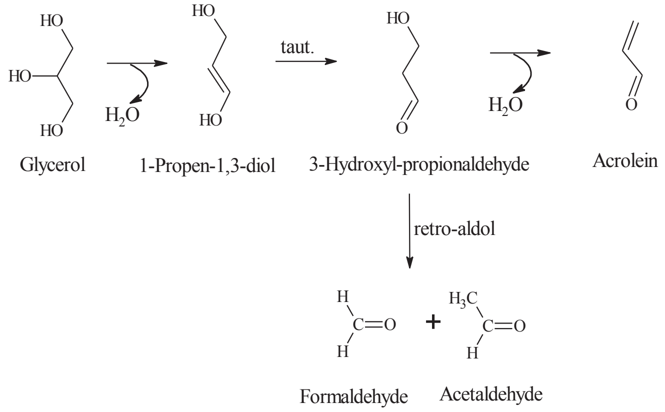

Fig. 1. The pyrolytic reactions of glycerol to produce formaldehyde, acetaldehyde and acrolein. Radical intermediates for steps involving loss of water are omitted for simplicity.

study on the formation of aldehydes in EC aerosol using a variety of devices and power levels.

It should be noted that PG can also decompose thermally, to propionaldehyde (Dai et al., 2004), however, in order to better compare to the previous studies mentioned above, which only reported formaldehyde, acetaldehyde and acrolein production, and to focus more on device dependence of their formation, we did not analyze for propionaldehyde in this study. Such analysis, as well as dependence on EC liquid solvent composition, is planned for future studies.

2. Methods

2.1. EC devices

In this study, five refillable “tank” based EC were studied:

- Device 1: Single top coil, 2.8 Ω,
- Device 2: Single bottom coil, 2.7 Ω,
- Device 3: Dual bottom coil, 2.8 Ω,
- Device 4: Single bottom coil, 2.2 Ω, and
- Device 5: Single bottom coil, 0.72 Ω.

All samples were commercial “tank” products (Hare, 2015) and were used according to the manufacturer's instructions. They all have similar functional parts: a tank which holds the liquid, a resistive heating wire (“coil”) to which voltage is applied to generate heat and aerosolize the liquid, a “wick” which can be silica string (Devices 1–3), poly-fill (Device 4) or cotton (Device 5), that transports the liquid in the tank to the coil, a mouth piece for inhalation, and a threaded connector to attach to and receive current from the power source. Device 1 was a CE4 “top-coil” tank-style (Vision, Shenzhen, China). Three separate devices were used in this study, and from the same manufacturer, all virtually identical save for some variations in coil resistance. The three devices used in this study were determined to have coil resistance of 2.2, 2.8 and 3.4 Ω (average was 2.8 Ω, with standard deviation of 0.5 Ω). In this device, the liquid is held inside a tank, and silica strings acting as wicks descend from a ceramic cup containing the coil into the liquid, which is fed to the coil through the wicks. “CE4” refers to the general design, using a ceramic coil cup, fourth version of this type of tank system. Air flow travels up through a center tube to under the coil, and then to the mouth. Adequate wetting of any EC coil depends on the ability of the wick to feed the liquid as fast as the coil vaporizes it. It should be noted that this style of atomizer is largely out of favor now in the vaping community, due to the difficulty of wicking with some liquids, and the propensity for dry-puff to occur. It should also be noted that this was the atomizer style chosen recently by previous researchers who reported high aldehyde and acrolein content of EC aerosol using 5 V or more (Jensen et al., 2015). Device 2 was a Protank 1 (KangerTech, Shenzhen, China) with a replaceable 2.7 Ω bottom single-coil-head. A single tank and three separate coils were used in this study. In this device the liquid is held in a tank and gravity fed to the coil, which is positioned at the bottom of the tank, through short silica wicking threads which the coil is wrapped around and oriented horizontally if the tank is held tip-up. It was expected that this design would allow more consistent wetting of the coil compared to Device 1. Device 3 was a Gladius (Innokin, Shenzhen, China) bottom coil tank system with a replaceable dual-coil-head and a total resistance of 2.8 Ω. A single tank and three unique coil-heads were used in this study. The overall design with respect to liquid feed is very similar to the Protank, but here there are two coils in parallel, at 5.6 Ω each, each wrapped horizontally around short silica wicks, stacked vertically on top of each other and across the central air-flow, which travels through a center tube to the mouth. The two coils in parallel have the effect of spreading the heat out evenly over the coils, compared to one coil when the same wattage is applied, assuming total resistance and all other factors are identical. Device 4 was bottom single coil Nautilus (Aspire USA, Kent, WA) with 2.2 Ω resistance. The overall design is visually similar to the Protank, but the replaceable coil-head is larger and the coil is vertically oriented, longer and of thicker gauge, and in contact with more wicking material (poly-fill). A single tank and three unique coil-heads were used in this study. Device 5 was a SubTank (KangerTech) with a 0.72 Ω bottom-coil-head. Since wattage is inversely proportional to coil resistance, reducing coil resistance will increase the wattage for a given battery voltage proportionally, allowing very high wattage from typical 3.7 V Li-ion batteries. The coil is vertically oriented, similar to the Nautilus coil-head, but the wicking material is cotton. A single atomizer was used with each device. In all cases, samples were collected from lowest power to highest power levels. All tanks were maintained at a minimum of 50% of the maximum liquid level. Where adjustment was possible for a device, airflow was set to maximum. Detailed images for the devices used in this study are available online (Google, 2015) and schematics of example top coil and bottom coil devices are given in supplemental materials Appendix A.

2.2. Sample collection

Puffing of devices was carried out using either a Cerulean SM450 (Milton Keyes, UK) or a KC Automation KC-5 (Richmond, VA) analytical smoking machine. The smoking regime was a puff every 30 s with 4-s duration and a volume of 55 mL collected using a “square” wave profile (Farsalinos et al., 2013). All devices were automatically activated at the start of each puff using an air power linear actuator attached to the battery. The button on each device was depressed during each puff. All devices were puffed with the tank held in a horizontal orientation. Between each puff block, devices were removed from the smoking machine to recorded the weight change. During the weighing process the devices were transported in a vertical orientation to allow for liquid equilibration. A puff block consisting of 25 puffs was performed and collected for each device and condition in duplicate, and this was repeated twice more with different units of the same device, three times total. Thus, each device and condition was averaged over 6 trials (N = 6). Batteries were fully charged before use, and the weight of each device was measured before and after each puff block. Devices were allowed to rest for least ten minutes between