



E-cigarettes as a source of toxic and potentially carcinogenic metals

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ABSTRACT

Background and aims: The popularity of electronic cigarette devices is growing worldwide. The health impact of e-cigarette use, however, remains unclear. E-cigarettes are marketed as a safer alternative to cigarettes. The aim of this research was the characterization and quantification of toxic metal concentrations in five, nationally popular brands of cig-a-like e-cigarettes.

Methods: We analyzed the cartomizer liquid in 10 cartomizer refills for each of five brands by Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

Results: All of the tested metals (cadmium, chromium, lead, manganese and nickel) were found in the e-liquids analyzed. Across all analyzed brands, mean (SD) concentrations ranged from 4.89 (0.893) to 1970 (1540) µg/L for lead, 53.9 (6.95) to 2110 (5220) µg/L for chromium and 58.7 (22.4) to 22,600 (24,400) µg/L for nickel. Manganese concentrations ranged from 28.7 (9.79) to 6910.2 (12,200) µg/L. We found marked variability in nickel and chromium concentration within and between brands, which may come from heating elements.

Conclusion: Additional research is needed to evaluate whether e-cigarettes represent a relevant exposure pathway for toxic metals in users.

1. Introduction

E-cigarettes are increasing in popularity in the United States with sales in 2015 exceeding \$3.5 billion (Herzog, 2015). There is great controversy surrounding e-cigarettes and some evidence showing that e-cigarettes are not harmless, although less so than cigarettes and may have long-term health implications for the user (Rom et al., 2015; Grana et al., 2014). Many of the active smokers who switch to e-cigarettes, and never smokers who start using them, do so in the belief that these devices are safer than combustible tobacco (Etter and Bullen, 2011; Goniewicz et al., 2013).

Cig-a-likes, the rechargeable or fully disposable devices commonly sold at convenience and liquor stores, are sometimes referred to as “first-generation” devices, implying that these e-cigarettes are waning in popularity (Lechner et al., 2015). We chose to analyze cig-a-likes because as of 2015, cig-a-likes still maintained a strong market share, despite falling in popularity compared to “second-generation” devices (Herzog and Gerberi, 2013). Surveys of e-cigarette users report that 99% of adult users are former or current smokers (Etter and Bullen, 2011; Etter, 2010). Over 80% of e-cigarette users are former tobacco

smokers (defined as no longer smoking any tobacco cigarettes) (Etter and Bullen, 2011; Piñeiro et al., 2016). In the US, e-cigarette use is increasing among teenagers who have never used tobacco (McCarthy, 2014, 2015; Wills et al., 2015; Gilreath et al., 2016).

Regulation of e-cigarettes varies across countries although at the time this research was conducted, cig-a-likes were unregulated in the US. Recently however, the US Food and Drug Administration (FDA) has announced new deeming regulations that bring e-cigarettes under the same regulations as tobacco (US Food and Drug Administration). Scheduled to come into effect as of August 2016, the rules require FDA approval for all e-cigarette products which entered the market after 2007. This move may have a substantial impact on the e-cigarette market and could potentially increase the market share of cig-a-like devices in the US, as many of these devices are produced by established tobacco companies who may be better positioned to afford the high cost of FDA product approval than smaller, independent device and e-liquid producers (Yandle et al., 2015). The European Union (EU) has also recently implemented regulations on e-cigarettes (Directive 2014/40/EU). These regulations include new labeling requirements and advertising restrictions.

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Table 1
Metal concentrations in five commercial brands of cig-a-like e-cigarettes ($\mu\text{g/L}$).

Brand	N	Cadmium			Chromium			Lead			Manganese			Nickel		
		Mean (SD)	Median	Range	Mean (SD)	Median	Range	Mean (SD)	Median	Range	Mean (SD)	Median	Range	Mean (SD)	Median	Range
Brand A ($\mu\text{g/L}$)	10	205 (318)	12.40	322–755	2110 (5220)	213	98.6–16,900	1970 (1450)	1630	500–4870	6910 (12,200)	918	541–31,500	22,600 (24,400)	15,400	2040–72,700
Brand B ($\mu\text{g/L}$)	10	1.17 (1.09)	0.796	0.470–4.11	788 (284)	726	306–1130	58.1 (79.4)	18.5	3.53–218	670 (283)	627	247–1200	13,400 (4540)	13,100	4560–20,500
Brand C ($\mu\text{g/L}$)	8	1.57 (1.30)	1.17	0.157–4.18	231 (71.6)	205	162–381	5.83 (1.80)	5.15	4.50–9.82	200 (33.9)	187	154–258	463 (132)	491	316–652
Brand D ($\mu\text{g/L}$)	10	0.982 (0.802)	0.502	0.249–2.23	76.1 (11.0)	75.6	60.2–92.7	4.89 (0.893)	4.98	3.17–5.89	41.50 (13.9)	44.4	11.8–65.5	58.7 (22.4)	58.1	13.7–85.4
Brand E ($\mu\text{g/L}$)	10	0.415 (0.38)	0.204	0.137–1.23	53.9 (6.95)	56.7	41.5–60.79	93.4 (80.5)	69.3	7.94–233	28.7 (9.79)	26.1	15.5–48.23	114 (49.3)	134	39.3–175
LOD ($\mu\text{g/L}$) [*]			0.04			0.1			0.02			0.08			0.1	
Intra-laboratory ICC	48×2		0.965			0.999			0.997			1.000			1.000	
Inter-laboratory ICC	4×2		0.997			0.993			0.997			0.988			0.988	

ICC: intraclass correlation coefficient. The intra-laboratory ICC was calculated from duplicate aliquots from the same e-cigarette liquid sample. Mean concentration was calculated by taking the mean of 2 duplicate samples from the same e-cigarette. The inter-laboratory ICC was calculated from duplicate analyses conducted in a subset of 4 e-cigarette liquid samples conducted at Graz University (Graz, Austria). *LOD are calculated to a 1:20 dilution factor.

Cig-a-like devices work by heating a liquid mixture of propylene glycol, glycerin, nicotine and flavorings. When heated with a metal coil, the mixture is aerosolized into a “vapor”, which is inhaled by the user. The commonly held belief among consumers of e-cigarettes is that they are a safer alternative to cigarettes (Goniewicz et al., 2013; Dockrell et al., 2013; Farsalinos et al., 2014). However, based on investigations including our own, there is strong evidence to suggest that these devices may be a source of toxic chemical exposure for users, particularly substances with known carcinogenic properties (Chervona et al., 2012; Cheng, 2014; Lerner et al., 2015; Tokar et al., 2011; Varlet et al., 2015; Barrington-Trimis et al., 2014).

Very little research has evaluated the potential of e-cigarettes to be a source of toxic metal exposure, including metals with known carcinogenic properties. To date, few published studies have investigated metal concentrations in US e-cigarette brands (Goniewicz et al., 2014; Williams et al., 2013). Goniewicz et al. investigated 12 Polish and British cig-a-like e-cigarettes and identified only nickel, cadmium and lead in cig-a-like aerosol, and in concentrations similar to that of a commercially available nicotine inhaler (Goniewicz et al., 2014). Concentrations ranged from 0.11 to 0.29 $\mu\text{g/e-cigarette}$ (150 puffs) for nickel and 0.03–0.57 $\mu\text{g/e-cigarette}$ for lead. That study did not report chromium or manganese in any brand. Williams et al. analyzed metal concentration in both liquid and aerosol and report the presence of nickel, chromium and lead, but not cadmium (Williams et al., 2013). Reported concentrations were 0.005 $\mu\text{g}/10$ puffs for nickel, 0.007 $\mu\text{g}/10$ puffs for chromium and 0.017 $\mu\text{g}/10$ puffs for lead (Williams et al., 2013).

The aim of this study was to analyze metal concentrations in the liquid of popular brands of e-cigarettes.

2. Materials and methods

We selected five popular brands of rechargeable “cig-a-like” devices available in the United States. The retail environment and sales of cig-a-likes are difficult to determine. Brands increase and decrease in popularity rapidly as cig-a-like manufacturers bring new products to market (Zhu et al., 2014). We chose five brands based on national market share. Three of the brands we tested comprised 71% of the market share of cig-a-likes in 2015 (Craver, 2015). Three of the brands are manufactured by tobacco companies and two are not, but all brands

are available nationally in the US at big-box retail outlets, convenience stores, and online. All brands contained nicotine in concentrations of approximately 1.6–1.8 mg/mL, as stated by the manufacturer on the cartridge packaging.

Cartridges from each brand were purchased at retail outlets or online. The liquid from 10 cartridges from each brand were analyzed. For each cartridge, we aimed to obtain enough liquid sample (approximately 400 μL) for two replicates. In the end we had a total of 48 liquid samples instead of 50 because two samples from Brand C did not yield enough liquid for analysis and those two samples were excluded. We only selected one flavor for each brand and flavor choice was determined by retail availability at the time of purchase. We found that total volume of liquid per cartridge varied significantly by brand and ranged from 300 to 600 μL . For this reason we chose not to measure per-cartridge metal content but instead report metal concentrations in $\mu\text{g/L}$, which allows for consistency in reporting across brands.

The end caps of each cartomizer were removed with standard pliers and the pad, free of the heating coil, was removed from the cartridge using polypropylene forceps. Pads were centrifuged for 10 min at 1540 RCF. Two aliquots of 250 μL were collected from each sample for Brand A, Brand B, Brand D and Brand E, and 150 μL for Brand C and diluted to 5 mL final volume with 1% HNO_3 and 0.5% HCl (Fisher Optima Trace Element Grade) in ultra-pure MilliQ water and vortexed prior to analysis. Cd, Cr, Pb, Mn, and Ni were analyzed using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500ce Octopole ICP-MS, Agilent Technologies, Santa Clara, USA). Method limits of detection (MLD) were calculated using procedural blanks and are reported in Table 1. Accuracy was successfully tested using NIST traceable Certified Reference Material TMDW-B (High Purity Standards, Charleston, SC). We estimated the intra-class correlation coefficient (ICC) for the two aliquots from the same sample (intra-laboratory ICC) and given the high reliability (Table 1), we calculated and used in the analysis the mean metal concentration of the two replicates for each e-cigarette liquid sample. We also conducted a duplicate analysis in a random subset of four e-cigarette liquid samples at the Trace Element Laboratory of the Institute of Chemistry Analytical Chemistry, Graz University (Graz, Austria), showing high comparability between laboratories (inter-laboratory ICC, Table 1).

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