

Does e-cigarette consumption cause passive vaping?

Abstract Electronic cigarette consumption ('vaping') is marketed as an alternative to conventional tobacco smoking. Technically, a mixture of chemicals containing carrier liquids, flavors, and optionally nicotine is vaporized and inhaled. The present study aims at the determination of the release of volatile organic compounds (VOC) and (ultra)fine particles (FP/UFP) from an e-cigarette under near-to-real-use conditions in an 8-m³ emission test chamber. Furthermore, the inhaled mixture is analyzed in small chambers. An increase in FP/UFP and VOC could be determined after the use of the e-cigarette. Prominent components in the gas-phase are 1,2-propanediol, 1,2,3-propanetriol, diacetyl, flavorings, and traces of nicotine. As a consequence, 'passive vaping' must be expected from the consumption of e-cigarettes. Furthermore, the inhaled aerosol undergoes changes in the human lung that is assumed to be attributed to deposition and evaporation.

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Key words: Electronic cigarette; Indoor air quality; Formaldehyde; Ultrafine particles; Propylene glycol; Third-hand smoke.

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Received for review 20 February 2012. Accepted for publication 28 May 2012.

Practical Implications

The consumption of e-cigarettes marks a new source for chemical and aerosol exposure in the indoor environment. To evaluate the impact of e-cigarettes on indoor air quality and to estimate the possible effect of passive vaping, information about the chemical characteristics of the released vapor is needed.

Introduction

Electronic cigarettes show a rapidly growing market share and are advertised as a healthier alternative to conventional smoking. These 'e-cigarettes' contain a small battery-driven heating unit that vaporizes a mixture of chemicals, the so-called 'liquids'. They usually contain flavors and carrier substances and may be purchased with and without nicotine. The nicotine content roughly differs between 0 and 20 mg/ml depending on the brand (Trehy et al., 2011). A common carrier of the 'liquids' is 1,2-propanediol (propylene glycol, PG) that leads to a visible fume during exhalation. This compound is also frequently used as a solvent in dosage formulations of aerosolized drug delivery systems such as pressurized metered-dose inhalers and nebulizers for the clinical practice (Montharu et al., 2010). However, the frequency of use is expected to be higher in case of e-cigarette vaping,

leading to a different exposure pattern. Propylene glycol is also a common humectant for tobacco cigarettes (Paschke et al., 2002). In contrast to conventional cigarettes, the released compounds are not generated from a combustion process (as a smoke) but by direct evaporation (as a vapor). For this reason, the term 'vaping' has been established among e-cigarette users as an analog to the conventional cigarette 'smoking' (Etter, 2010).

A recent study reports adverse physiological effects after the short-term use of e-cigarettes (Vardavas et al., 2011). This effect may be attributed to propylene glycol that is known to cause upper airway irritations (Wieslander et al., 2001). However, a comprehensive exposure assessment that compares the nicotine intake from e-cigarettes and conventional cigarettes – which also considers the impact of the carrier substances – is not available at the present state. Furthermore, the release of the organic compounds from the 'liquids' and

the release of particles into the indoor environment are still mostly unknown. In contrast, the impact of environmental tobacco smoke from conventional smoking on the indoor air quality has been intensively researched in the past decade. Numerous studies report the release of particulate matter (Nazaroff and Klepeis, 2003) and organic compounds such as formaldehyde, from the combustion of tobacco products (Baek and Jenkins, 2004; Baker, 2006; Paschke et al., 2002). These scientific findings led to a ban on smoking in public buildings and restaurants in many countries. This ban had a positive influence on the indoor air quality in these buildings (Bohac et al., 2010; Gleich et al., 2011).

Beyond indoor climate, airflow conditions, room size, and number of e-cigarette users, many other parameters have the potential to affect 'passive vaping'. The concentrations of the exhaled compounds during e-cigarette consumption can be expected to differ with the composition of the applied 'liquids', the type of e-cigarette in use, the age of the e-cigarette (e.g., owing to remains of previous 'liquids'), length of the puff, and interval between the puffs. Moreover, the composition of the exhaled air will be affected by age, sex, activity, health status, and diet of the user (Riess et al., 2010).

Another important aspect in the future discussion about e-cigarettes will be the effect of 'third-hand smoke' that mainly describes human exposure against residues of smoking on clothes, furniture, and other indoor surfaces (Matt et al., 2011). In case of e-cigarettes, the solvent of the 'liquids' may remain on available surfaces and be a source for the contamination of residents. Even more important might be the accidental spilling of 'liquids' that can lead to unintended uptake of nicotine by skin permeation – an effect that is intentionally used for nicotine patches (Hammer et al., 2011). It can be assumed that the health impact of e-cigarette use is mainly influenced by the safety and quality of the applied 'liquids'.

The present study provides first indications about the entry of volatile organic compounds (VOCs) and ultrafine particles into the indoor environment connected with the use of electronic cigarettes. One measurement was performed in a full-scale emission test chamber with one e-cigarette and different 'liquids'. Additional small-scale chamber measurements were performed to identify the effect of aerosol aging and the impact of different e-cigarette types. The experiments aim at the identification of the released compounds under near-to-real-use conditions to estimate the effect of 'passive vaping'.

Material and methods

Large-scale vaping/smoking experiment

The experiment was performed in an 8-m³ stainless-steel emission test chamber. This chamber was oper-

ated at 23°C and 50% relative humidity at an air exchange rate of 0.3/h. The formaldehyde concentration in the chamber was continuously recorded every 30 s by an AL4021 formaldehyde auto analyzer (AeroLaser). A fast mobility particle sizer (FMPS; TSI Inc., Shoreview, MN, USA) recorded the particle number concentration of fine and ultrafine particles (FP/UFP) in the size range between 5.6 and 560 nm at 1 Hz in 32 channels.

Before the experiment and after each smoking event, 3 l of chamber air was pumped (200 ml/min) through stainless-steel tubes filled with 300 mg Tenax TA. The tubes were analyzed via thermal desorption (UltraUnity 2; Markes Int., Llantrisant, UK) and gas chromatography (6890 Series GC System; Agilent, Santa Clara, CA, USA; HP5MS 60 m × 250 μm × 0.3 μm column) coupled with mass spectrometry (5973N MSD; Agilent) according to ISO 16000-6. In parallel, lower aldehydes (formaldehyde, acetaldehyde, etc.) were collected using silica gel cartridges containing 2,4-dinitrophenylhydrazine (DNPH). The cartridges were analyzed according to ISO 16000-3 using high-performance liquid chromatography coupled with a variable wavelength detector (HPLC 1200 Infinity; Agilent).

A volunteering smoker took a seat in the chamber, and the chamber blank was measured after 20 min of conditioning. The e-cigarette was then filled with an apple-flavored nicotine-free 'liquid' (Liquid 1) outside of the chamber and given to the test person through a sampling port. The person took six deep-lung puffs (puff length ~ 3 s) with a delay of 60 s between each puff. The air sampling on Tenax TA tubes started at puff 4 and lasted 15 min. This procedure was performed for another two 'liquids', Liquid 2 and Liquid 3 (see Table 1).

After the e-cigarette was removed from the chamber, a conventional tobacco cigarette was lit outside the chamber and given to the test person. The sampling procedure was identical to the e-cigarette measurement.

For the determination of the feasible puff length, the mouthpiece and the wick (see Figure 1) were removed from the e-cigarette and the temperature of the heating coil was measured via thermography (ThermaCAM B20; FLIR Systems, Wilsonville, OR, USA) during

Table 1 Characteristics of the 'liquids'

| Sample | Flavor | Main aroma compound | Nicotine content ^a |
|------------------------|---------|---------------------------------|-------------------------------|
| Liquid 1 | Apple | 3-Methylbutyl-3-methylbutanoate | 0 mg/ml |
| Liquid 2 | Apple | 3-Methylbutyl-3-methylbutanoate | 18 mg/ml |
| Liquid 3 | Tobacco | Ethyl maltol | 18 mg/ml |
| Conventional cigarette | – | – | 0.8 mg/cigarette |

^aAs stated by the manufacturer. [Correction added on 6 August 2012, after first online publication: Nicotine content for Liquid 2 and Liquid 3 changed from 1.8 mg/ml to 18 mg/ml.]

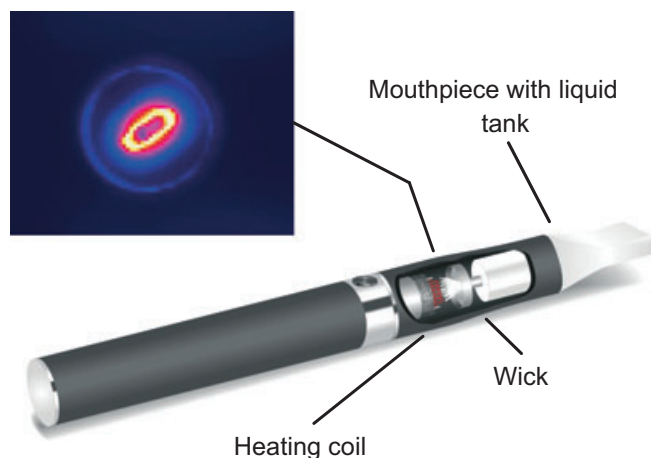


Fig. 1 Scheme of the tested e-cigarette A. The thermographic image shows the temperature distribution of the heating unit without ‘liquid’ ($> 350^{\circ}\text{C}$ in the center)

heat-up. The time-resolved analysis showed an interval of 3 s between start of the cigarette and reaching stable temperature conditions. The puff length was equally increased for e-cigarette and tobacco cigarette, even though the length of the puff was approximately 1 s longer than specified in ISO 3308 (2000). The puff interval (60 s) was selected according to ISO 3308. The number of puffs (10 in ISO 3308) had to be adapted to the new smoking conditions because the tobacco cigarette was depleted after six puffs.

Vapor analysis

An aerosol aging experiment was performed in a 10-l glass emission test chamber. The chamber is double walled and is temperature controlled by water. The air in the chamber is mixed by a small fan. The e-cigarette was connected to the inlet, and a pump was used to produce a slight underpressure that transfers the aerosol directly into the chamber. The e-cigarette was operated for 3 s. The aerosol was aged in the chamber for 1, 3, 5, 7, and 10 min at 37°C . Additionally, the aerosol was aged 5 min at 23, 37, and 50°C . Then, the FMPS (sample flow rate of 8 l/min) was connected to the chamber, and the chamber inlet was equipped with a HEPA filter.

Analysis of VOCs in exhaled breath

After measuring the VOC chamber blank, an e-cigarette consumer was asked to exhale one e-cigarette

Table 2 Characteristics of the tested e-cigarettes

| Sample | Casing | Delivery system | Comparative price |
|---------------|------------------------|-----------------|-------------------|
| e-Cigarette A | Stainless steel/rubber | Tank | High (>35 Euro) |
| e-Cigarette B | Stainless steel | Cotton | Medium |
| e-Cigarette C | Stainless steel | Tank | Low (<25 Euro) |

puff into the 10-l glass chamber. The VOCs within the chamber were then determined by GC/MS after sampling on Tenax TA tubes (6L, 150 ml/min).

Measurement with three different e-cigarettes

Three different types of e-cigarettes (see Table 2) were filled with ‘liquid’ from the same stock (Liquid 1). The cigarette was operated for 3 s. The vapor from the e-cigarettes was transferred into the 10-l glass chamber using a pump. The chamber was set to 37°C and an air exchange rate of 3/h. Directly after injection of the vapor, sampling on Tenax TA was performed for 60 min (100 ml/min) and sampling on DNPH was performed for 200 min (120 ml/min). Between each measurement, the chamber was heated to 60°C for 24 h at maximum air exchange rate (6/h). The measured concentration c_S ($\mu\text{g}/\text{m}^3$) is converted into the released mass per puff MPP ($\mu\text{g}/\text{puff}$) according to Equation 1 using the sample volume V_S (m^3), the number of puffs n (puff), and the ratio between sample flow \dot{V}_S (m^3/h) and chamber exhaust flow \dot{V}_C (m^3/h). Additionally, the value is corrected for the expected exponential decay of the concentration because of the air exchange rate k (/h).

$$\text{MPP} = \frac{c_S}{n} \cdot V_S \cdot \frac{\dot{V}_C}{\dot{V}_S} \cdot \frac{\int_0^{\infty} e^{-k \cdot t} dt}{\int_0^t e^{-k \cdot t} dt} = \frac{c_S}{n} \cdot V_S \cdot \frac{\dot{V}_C}{\dot{V}_S} \cdot \frac{1}{1 - e^{-k \cdot t}} \quad (1)$$

Descriptions of the performed experiments as well as the measured climatic conditions during measurement are summarized in Table 3.

Results and discussion

Emission of volatile organic compounds

Electronic cigarettes use a completely different principle of operation compared to tobacco cigarettes. The ‘liquid’ is vaporized and because of the thermodynamic properties of 1,2-propanediol ($K_p = 188^{\circ}\text{C}$, $\Delta H_v = 64.5$ kJ/mol at 298.15 K) (Verevkin, 2004), the heat from the coil (see Figure 1) is led off, which avoids pyrolysis. In contrast, conventional cigarettes release numerous compounds into the indoor environment. Paschke et al. (2002) listed hundreds of ingredients in tobacco cigarettes that form volatile combustion products. In Table 4, the 20 compounds with the highest concentrations in the 8-m^3 chamber air are summarized. During operation of the e-cigarette, the carrier substance of the ‘liquids’, 1,2-propanediol, was detected in the chamber atmosphere but the concentration was below the limit of determination. In contrast, a high concentration of 1,2-propanediol was observed for smoking of the conventional cigarette. The compound is known to be pyro-

Table 3 Description of the performed experiments

| Experiment | Chamber | T (°C) ^a | RH (%) ^a | e-Cig. | 'Liquid' | Smoker | Analytics |
|------------------------|----------------------------------|--|--|-------------|----------|--------|---|
| Large-scale experiment | 8-m ³ stainless steel | 24.1 ± 1.1 | 44.5 ± 8.2 | A | 1–3 | Yes | Fast mobility particle sizer (FMPS), AeroLaser, Tenax, DNPH |
| Vapor analysis/aging | 10-l glass | 22.7 ± 0.1 37.1 ± 0.2 49.9 ± 0.1 | 36.9 ± 0.5 18.9 ± 0.6 11.0 ± 0.6 | A | 1 | No | FMPS |
| Exhaled breath | 10-l glass | 37.0 ± 0.2 | 27.2 ± 4.3 | A | 1 | Yes | Tenax |
| Three e-cigarettes | 10-l glass | 36.8 ± 0.2 37.1 ± 0.2 37.1 ± 0.2 | 20.2 ± 0.6 18.2 ± 0.6 17.7 ± 0.6 | A B C | 1 | No | Tenax, DNPH |

^aThese values provide the measured mean climatic conditions (measuring interval: 1 min) and the standard deviations during performing the experiments.

Table 4 Concentrations ($\mu\text{g}/\text{m}^3$) of selected compounds during the 8-m³ emission test chamber measurement of e-cigarette A and conventional cigarette using Tenax TA and DNPH

| Compounds | CAS | Participant blank | E-cigarette | | | Conventional cigarette |
|---------------------------------|-----------|-------------------|-------------|----------|----------|------------------------|
| | | | Liquid 1 | Liquid 2 | Liquid 3 | |
| 1,2-Propanediol | 57-55-6 | <1 | <1 | <1 | <1 | 112 |
| 1-Hydroxy-2-propanone | 116-09-6 | <1 | <1 | <1 | <1 | 62 |
| 2,3-Butanedione | 431-03-8 | <1 | <1 | <1 | <1 | 21 |
| 2,5-Dimethylfuran | 625-86-5 | <1 | <1 | <1 | <1 | 5 |
| 2-Butanone (MEK) | 78-93-3 | <1 | 2 | 2 | 2 | 19 |
| 2-Furaldehyde | 98-01-1 | <1 | <1 | <1 | <1 | 21 |
| 2-Methylfuran | 534-22-5 | <1 | <1 | <1 | <1 | 19 |
| 3-Ethenyl-pyridine ^a | 1121-55-7 | <1 | <1 | <1 | <1 | 24 |
| Acetic acid | 64-19-7 | <1 | 11 | 13 | 14 | 68 |
| Acetone | 67-64-1 | <1 | 17 | 18 | 25 | 64 |
| Benzene | 71-43-2 | <1 | <1 | <1 | <1 | 22 |
| Isoprene | 78-79-5 | 8 | 6 | 7 | 10 | 135 |
| Limonene | 5989-27-5 | <1 | <1 | <1 | <1 | 21 |
| m,p-Xylene | 1330-20-7 | <1 | <1 | <1 | <1 | 18 |
| Phenol | 108-95-2 | <1 | <1 | <1 | <1 | 15 |
| Pyrrrole | 109-97-7 | <1 | <1 | <1 | <1 | 61 |
| Toluene | 108-88-3 | <1 | <1 | <1 | <1 | 44 |
| Formaldehyde ^b | 50-00-0 | <1 | 8 | 11 | 16 | 86 |
| Acetaldehyde ^b | 75-07-0 | <1 | 2 | 2 | 3 | 119 |
| Propanal ^b | 123-38-6 | <0.2 | <0.2 | <0.2 | <0.2 | 12 |

^aQuantified on the basis of toluene response.

^bDNPH method.

lyzed to acetaldehyde and acetone during smoking (Paschke, 2002).

Ohta et al. (2011) proposed the formation of formaldehyde, acetaldehyde, and methylglyoxal in the e-cigarette because of the oxidation of propylene glycol during contact with the active heating coil. However, continuous monitoring only showed a slight increase in the formaldehyde concentration in the 8-m³ emission test chamber before and during the consumption of the three 'liquids' (see Table 4 and Figure 2). This might be caused by the person in the chamber itself, because people are known to exhale formaldehyde in low amounts (Riess et al., 2010) and the increase was already observed during the conditioning phase (Figure 2). Furthermore, the release of formaldehyde was also below the limit of detection in the small-scale experiments. The expected rise of the formaldehyde

concentration in the chamber from smoking a conventional cigarette with a peak value of 114 ppb is shown in Figure 2. Other indoor pollutants of special interest, such as benzene, were only detected during the tobacco smoking experiment. The rising concentrations of acetic acid and acetone during e-cigarette operation may also be attributed to the metabolism of the consumer.

Although 1,2-propanediol was detected in traces only in the 8-m³ chamber during the consumption of e-cigarettes, this compound must be released owing to the visible fume in the exhaled breath. To determine the VOC composition in the breath gas directly, an e-cigarette smoker exhaled into a 10-l glass chamber. The identified chemical species are shown in Figure 3. The experiment revealed a high amount of 1,2-propanediol in the exhaled air. Other main components were the

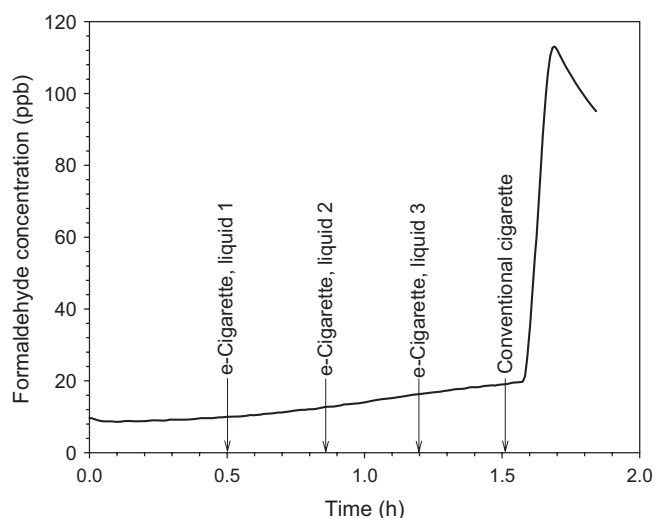


Fig. 2 Formaldehyde concentration in the 8-m³ test chamber during consumption of e-cigarettes (Liquids 1–3) and one conventional cigarette

carrier substance 1,2,3-propanetriol, the flavoring source diacetin as well as traces of apple oil (3-methylbutyl-3-methylbutanoate) and nicotine. The fact that these compounds were not detectable during the 8-m³ emission test chamber measurement is assumed to be caused by the short usage (6 min per ‘liquid’) and sink effects of the chamber for the very polar 1,2-propanediol.

Regarding the variability of e-cigarettes, the VOC emission strength seems to differ with different types of e-cigarettes (Table 5). While the e-cigarettes A and C have similar emission patterns, the emission from e-cigarette B is significantly higher. Formaldehyde was not detected during any measurement. With e-cigarette C, almost three times more propylene glycol is released per puff. This deviation is assumed to be

caused by the ‘liquid’ supply technique. In case of e-cigarettes A and C, the ‘liquid’ is stored in a tank, while e-cigarette B features a cotton unit that is drenched with the ‘liquid’. However, a general correlation between emission strength and ‘liquid’ supply technique (tank or cotton) is not possible from this limited data set. The effect of other systems, such as underpressure-activated e-cigarettes, was not determined in this study and is an important topic for further research.

Aerosol release from the e-cigarette

The airborne particles being related to the e-cigarette experiment are assumed to be formed from supersaturated 1,2-propanediol vapor. In contrast to the conventional cigarette, which continuously emits particles from the combustion process itself, the e-cigarette aerosol is solely released during exhalation. The e-cigarette aerosol measured in the 8-m³ chamber is bimodal: one maximum is found in the range of 30 nm and one in the range of 100 nm (see Figure 4a). During the ongoing experiment, the ultrafine particle mode increased. The particles in the higher mode are assumed to be evaporated or deposited in the human lung. Because of the high vapor pressure of 1,2-propanediol ($p_s = 17.36$ Pa at 298.15 K) (Verevkin, 2004), the dynamics of the aerosol is expected to be fast. For comparison, the particle size distribution of the conventional cigarette provides a single mode with a maximum at 100 nm and a higher total number concentration (see Figure 4b).

For characterization of the e-cigarette aerosol, it was passed directly from the mouthpiece into a 10-l glass emission test chamber. Then, it was aged for 5 min at 23, 37, and 50°C, respectively. From Figure 5a, it is obvious

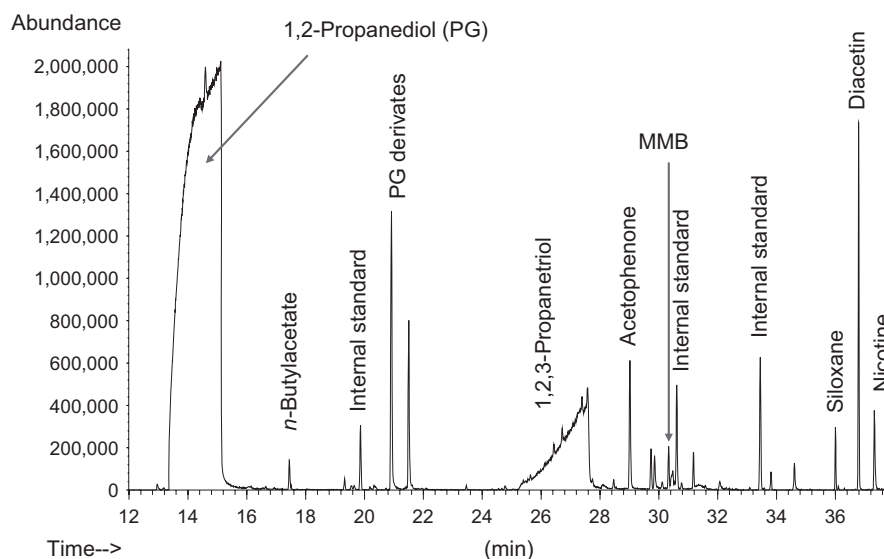


Fig. 3 Gas chromatogram of one exhaled e-cigarette puff (Liquid 2) in a 10-l glass chamber (sampled on Tenax TA, 3 l sampling volume) (MMB = 3-methylbutyl-3-methylbutanoate; PG = propylene glycol)

Table 5 Comparison of the release of volatile organic compound for a number of selected compounds from three types of e-cigarettes A-C (one puff, 3 s) in a 10-l glass chamber using Tenax TA and DNPH

| Compound | Concentration ($\mu\text{g}/\text{m}^3$) | | | Estimated mass per puff ($\mu\text{g}/\text{puff}$) ^a | | |
|---------------------------------|--|---------|--------|--|-------|-------|
| | A | B | C | A | B | C |
| 1,2-Propanediol | 53 000 | 175 000 | 64 000 | 1673 | 5525 | 2021 |
| 1,2,3-Propanetriol | 326 | 477 | 161 | 10 | 15 | 5 |
| 3-Methylbutyl-3-methylbutanoate | 3 | 35 | 10 | 0.1 | 1.1 | 0.3 |
| Diacetin | 2 | 1 | 1 | 0.06 | 0.03 | 0.03 |
| Triacetin | <1 | <1 | <1 | <0.03 | <0.03 | <0.03 |
| Nicotine | 7 | 7 | 4 | 0.2 | 0.2 | 0.1 |
| Formaldehyde ^b | <2 | <2 | <2 | <0.25 | <0.25 | <0.25 |
| Acetaldehyde ^b | <1 | <1 | <1 | <0.13 | <0.13 | <0.13 |
| Propanal ^b | <1 | <1 | <1 | <0.13 | <0.13 | <0.13 |

^aThe conversion factors based on the sample volume, the sample flow, and the exponential decay of the concentration (see Equation 1).

^bDNPH method.

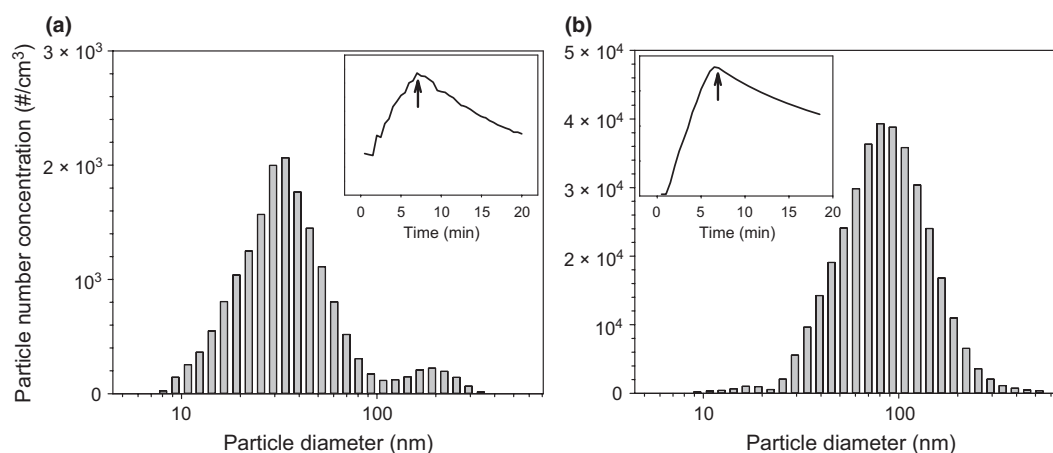


Fig. 4 (a) Aerosol size distribution during consumption of an e-cigarette in the 8-m³ chamber. (b) Aerosol size distribution during consumption of a conventional cigarette in the 8-m³ chamber. The arrows in the insets of (a) and (b) indicate the actual time in concentration development

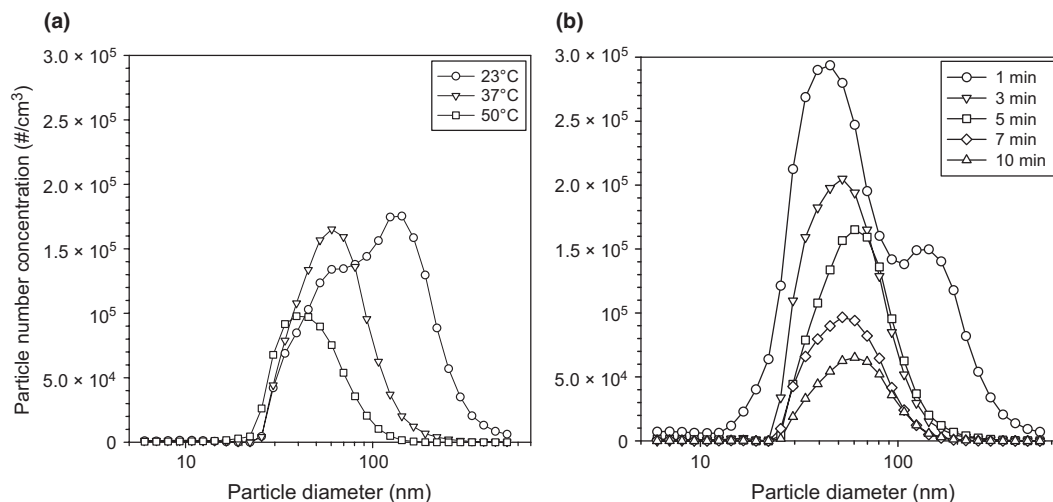


Fig. 5 Aerosol size distributions of aged e-cigarette aerosols in a 10-l glass chamber. The aerosol was aged for 5 min at different temperatures (a) and for different times at 37°C (b)

that because of increasing temperature, the aerosol shifts from a bimodal size distribution with maxima at 60 and 100 nm into a single-mode distribution with a maximum

at 45 nm. Figure 5b demonstrates the effect of aging at 37°C. Between 1 and 3 min, the higher mode at 100 nm disappeared and a single-mode aerosol with a maximum

at 45 nm is left. This ‘shrinking’ of the particles can be attributed to the evaporation of the particles under ideal conditions. However, in the real indoor environment, the present airborne particles might affect aging, for example, owing to coagulation. The inlet air of the large-chamber experiment was free of particles, and thus, the experimental results in both chambers are conclusive. In total, these findings prove that the influence of the e-cigarette on the indoor air particle concentration cannot be determined solely from direct aerosol sampling at the source. The dynamics and changes of the aerosol size distribution resulting from the dwell time in the human lung must be considered.

Conclusions

The consumption of e-cigarettes causes emissions of aerosols and VOCs, such as 1,2-propanediol, flavoring substances, and nicotine, into indoor air. During inhalation of e-cigarette vapor, the aerosol size distribution alters in the human lung and leads to an exhalation of smaller particles. This effect is caused

by the evaporation of the liquid particles in the lung and also in the environment after exhalation. The quantity of the inhaled vapor could be observed to depend on the ‘liquid’ delivery system of the e-cigarette in use.

Overall, the e-cigarette is a new source of VOCs and ultrafine/fine particles in the indoor environment. Therefore, the question of ‘passive vaping’ can be answered in the affirmative. However, with regard to a health-related evaluation of e-cigarette consumption, the impact of vapor inhalation into the human lung should be of primary concern.

Acknowledgements

The authors are grateful to Manuela Lingnau for preparing the e-cigarette scheme and Friedrich Schlüter for providing the thermographic image. The authors especially like to thank Frank Fuhrmann for stimulating the interest in the e-cigarette topic. This work was financially supported by internal Fraunhofer WKI research funds.

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