

Dynamic Imaging and Characterization of Volatile Aerosols in E-Cigarette Emissions Using Deep Learning-Based Holographic Microscopy

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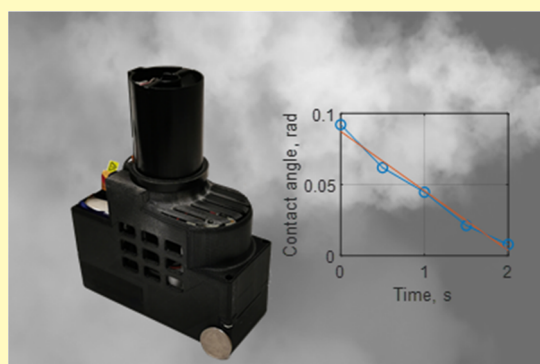
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Supporting Information

ABSTRACT: Various volatile aerosols have been associated with adverse health effects; however, characterization of these aerosols is challenging due to their dynamic nature. Here, we present a method that directly measures the volatility of particulate matter (PM) using computational microscopy and deep learning. This method was applied to aerosols generated by electronic cigarettes (e-cigs), which vaporize a liquid mixture (e-liquid) that mainly consists of propylene glycol (PG), vegetable glycerin (VG), nicotine, and flavoring compounds. E-cig-generated aerosols were recorded by a field-portable computational microscope, using an impaction-based air sampler. A lensless digital holographic microscope inside this mobile device continuously records the inline holograms of the collected particles. A deep learning-based algorithm is used to automatically reconstruct the microscopic images of e-cig-generated particles from their holograms and rapidly quantify their volatility. To evaluate the effects of e-liquid composition on aerosol dynamics, we measured the volatility of the particles generated by flavorless, nicotine-free e-liquids with various PG/VG volumetric ratios, revealing a negative correlation between the particles' volatility and the volumetric ratio of VG in the e-liquid. For a given PG/VG composition, the addition of nicotine dominated the evaporation dynamics of the e-cig aerosol and the aforementioned negative correlation was no longer observed. We also revealed that flavoring additives in e-liquids significantly decrease the volatility of e-cig aerosol. The presented holographic volatility measurement technique and the associated mobile device might provide new insights on the volatility of e-cig-generated particles and can be applied to characterize various volatile PM.

KEYWORDS: computational microscopy, digital holography, aerosol detection, volatility characterization, electronic cigarettes



Exposure to particulate matter (PM) has been associated with various adverse health effects in epidemiological studies;^{1,2} some of these particles contain a large fraction of volatile or semivolatile materials, such as emissions generated by cooking,³ vehicles,⁴ and usage of tobacco products.^{5,6} Since the particle dynamics-related information derived from non-volatile aerosols cannot be applied to volatile or semivolatile emission sources, there is a need to better understand and measure PM volatility and related dynamic behavior for assessing their exposure and potential health impact.

The dynamic evaporation of a volatile particle raised enduring research interest that can be traced back to the 19th century.⁷ Thermal denuders were used as a potential candidate, providing inline saturation pressure measurements, especially in aerosol studies.^{4,8,9} A thermal denuder compares particle size distribution and concentration of a cloud of volatile particles before and after they evaporate under precise control of the heating temperature and time. Thermal dynamic models are used to map differences in particle number and size distribution to the physical parameters of volatile particles. Unfortunately, such models are still missing for aerosols that

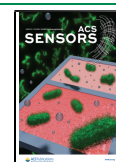
have complex chemical compositions.¹⁰ Another way of measuring a particle's volatility is to image its geometrical/structural change when it is sitting on a transparent substrate.^{7,11} However, these earlier methods are used to measure relatively large particles that have a footprint on the order of square millimeters and suffer from extremely low throughput.

Here, we report a high-throughput volatile particle measurement system that is based on computational inline holography and deep learning (Figure 1), which was applied for dynamic imaging and characterization of volatile aerosols created by electronic cigarettes (e-cigs). An impactor-based portable air quality monitoring device¹² was used to sample aerosols onto a

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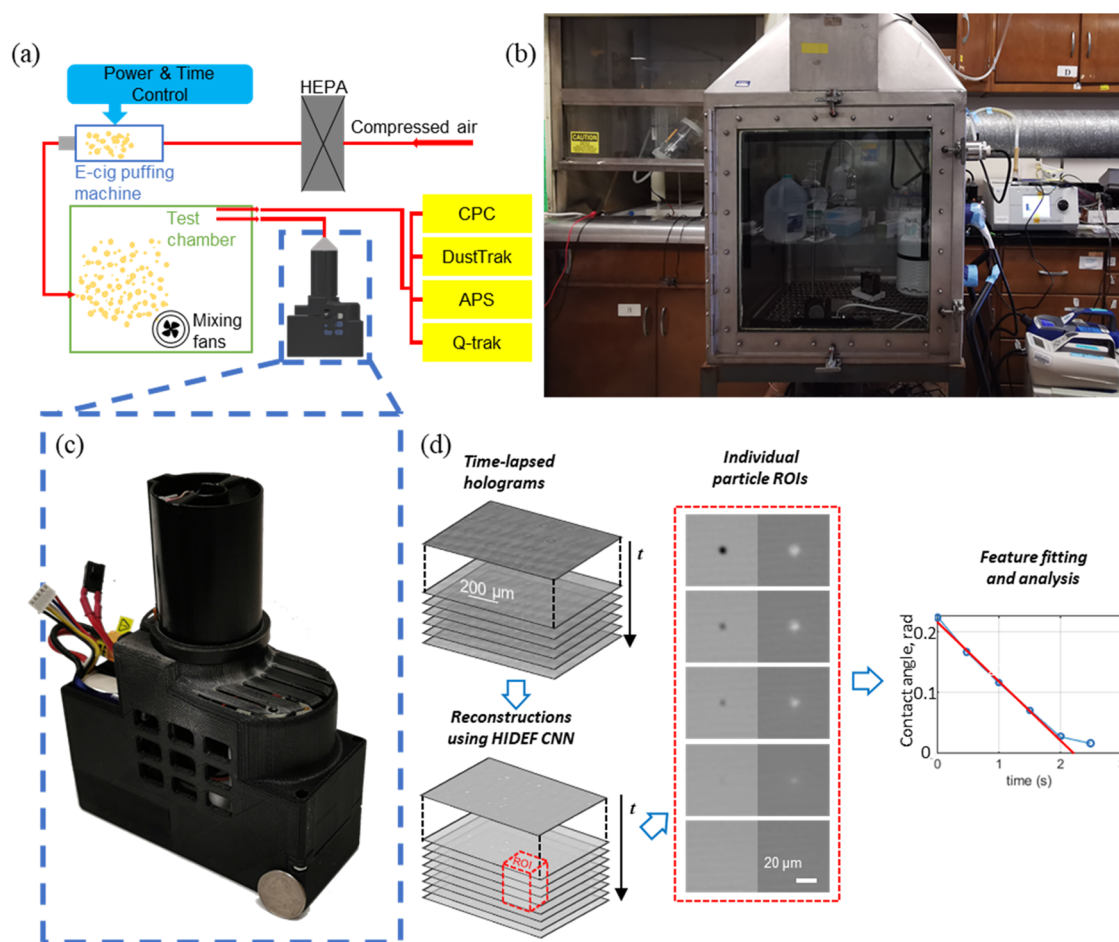


Figure 1. Computational imaging system for directly measuring the volatility of e-cig-generated aerosols. A customized puffing machine was used to simulate human puffing of e-cig. Puffing-generated e-cig aerosols were sampled by lens-free computational microscopy, and the volatility of each captured particle was quantified. (a) Schematic drawing of the puffing machine and the computational imaging system. (b) Photograph of the presented system. (c) Photograph of the computational imaging system, c-Air. (d) Image processing pipeline for measuring a dynamic particle's volatility.

transparent sticky sampling pad, at a throughput of 13 L/min. Time-lapsed inline holograms of the sampled particles (typically more than 10000 particles per experiment) were recorded at 2 frames per second (fps) across a field-of-view of 4 mm². A deep neural network¹³ was used to reconstruct the acquired inline holograms, revealing the phase and amplitude image of each particle as a function of time, which was further processed to measure its volatility. This system was applied to characterize e-cig-generated aerosols, which are highly dynamic^{14–16} due to the volatile materials used in e-cig liquid mixtures (e-liquids).

E-cigs have gained worldwide attention, primarily due to their unprecedented popularity among never-smoking adolescents and young adults over the last decade.^{17–20} The use of an e-cig (or vaping) generates an inhalable aerosol by heating and vaporizing an e-liquid, which typically uses propylene glycol (PG) and vegetable glycerin (VG) as the solvents to dilute nicotine and flavoring compounds. Many previous studies have reported the potential adverse health effects and toxicity of e-cig aerosols.^{21–26} The recent outbreak of e-cig or vaping product use-associated lung injury (EVALI) has particularly raised public health concerns.^{27–29} Previously, Li et al.³⁰ reported that the PG/VG ratio and the addition of nicotine in the e-liquid changed particle loss rates based on the slope of log-normalized PM concentrations, suggesting that evaporative

properties may be linked to the constituents of the e-liquid. However, this traditional method was relatively time-consuming and indirect since the particle loss rate estimation could not directly provide the volatility information. Other indirect methods for estimation of particles' volatility were also reported in the literature.^{31,32}

We used a holographic mobile imaging device to directly measure the volatility of individual aerosols generated by different e-liquid compositions using a customized puffing machine to validate the effects of e-liquid composition on aerosol volatility at the microscale. Our experimental results revealed a negative correlation between the e-cig aerosol's volatility and the volumetric ratio of VG in nicotine-free e-liquids. The addition of other chemicals, e.g., nicotine and flavoring chemicals, introduced significant changes in the observed volatility patterns. Nicotine was found to dominate the evaporation dynamics of e-cig emission and overwhelm the impact of VG volumetric ratio. The addition of flavoring chemicals into e-liquids also significantly decreased the volatility of e-cig-generated aerosols. The presented holographic volatility measurement technique with its mobile interface provides direct and high-throughput quantification of the volatility of e-cig-generated aerosols and can be broadly applied to rapidly characterize various volatile PM.

MATERIALS AND METHODS

E-Cigarette Sample Preparation. In this study, both homemade and commercially available e-liquids were tested. Homemade e-liquids were prepared from individual chemicals, including PG ($\text{C}_3\text{H}_8\text{O}_2$, $\geq 99.5\%$, Sigma-Aldrich), VG ($\text{C}_5\text{H}_8\text{O}_3$, $\geq 99.5\%$, Sigma-Aldrich), and nicotine ($\text{C}_{10}\text{H}_{14}\text{N}_2$, $\geq 99\%$, Sigma-Aldrich). Homemade e-liquids with five different PG/VG ratios (100/0, 70/30, 50/50, 30/70, and 0/100) and three different nicotine levels (0, 1.2, and 2.4% mass concentration) were prepared and tested. To ensure the quality of e-liquids, all of the studied e-liquids were well mixed and prepared within 7 days before each experiment. Commercial e-liquids were purchased from VaporVapes Inc. (Sand City, CA), including nicotine-free, flavorless e-liquid with five different PG/VG ratios, and three flavored, nicotine-free e-liquid with a PG/VG ratio of 50/50 (tobacco, menthol, and strawberry flavor).

Aerosol Generation and Characterization. To investigate the impact of e-liquid composition on e-cig-generated aerosol volatility, we created aerosols using a customized puffing machine mimicking human vaping (Figure 1a,b). The puffing machine was composed of an acrylic holder, an e-cig device with a refillable tank, a power source, and an Arduino UNO R3 microcontroller. The e-cig device used in the study was the Vapor-fi Volt II Hybrid Tank.^{30,33,34} In addition to this e-cig device/tank, we also investigated the impact of another e-cig device on volatility constant by measuring the emissions of a pod-type e-cig (JUUL, Juul Labs Inc.); this comparison between the two e-cig devices is reported in Supporting Information. The e-cig tank (Vapor-fi Volt II Hybrid Tank) was equipped with a $0.5\ \Omega$ heating coil and powered at 15 W (i.e., 6 V and 2.5 A). Clean air with a flow rate of 1 L/min was constantly fed into the puffing machine to transfer all generated aerosols into a test chamber. The test chamber used in this study has a volume of $0.46\ \text{m}^3$. The air exchange rate of the chamber was maintained at $1\ \text{h}^{-1}$ (i.e., the air inside the chamber gets replaced once every hour). The dilution ratio of the chamber is approximately 6900:1.³⁰ The temperature and relative humidity inside the test chamber were measured by an indoor air quality monitor (Q-Trak 7575, TSI Inc.) and were maintained at $50 \pm 10\%$ and $25.0 \pm 2.0\ ^\circ\text{C}$, respectively. A portable condensation particle counter (CPC 3007, TSI Inc.) was used to measure particle number concentration (PNC). To ensure data quality, the background PNC inside the chamber was kept at <100 particles/ cm^3 before each sampling session. An Aerodynamic Particle Sizer (APS 3321, TSI Inc.) was used to measure the particle size distribution ranging from 0.5 to $19.8\ \mu\text{m}$. A DustTrak II Aerosol Monitor (DustTrak 8532, TSI Inc.) was used to measure $\text{PM}_{2.5}$ concentrations.

Time-Lapsed Imaging of e-cig-Generated Aerosols. The e-cig-generated aerosol was imaged with a holographic PM monitoring device (termed c-Air;^{12,35,36} a photograph of the device is shown in Figure 1c) which was used to quantify its volatility, particle by particle. The c-Air device was powered by a lithium polymer (Li-po) battery (Turnigy Nano-tech 1000 mAh 4S 45–90C Li-po pack) and controlled by a Raspberry Pi Zero W single-board computer. The device was connected to the e-cig test chamber through a $1/4$ in. inner diameter Tygon tube. A miniaturized vacuum pump (M00198, GTEK Automation) inside c-Air-sampled aerosols from the tubing through a disposable impactor at 13 L/min, where the aerosols within the air stream landed on a sticky transparent coverslip. The c-Air device used a vertical-cavity surface-emitting laser (VCSEL) diode (OPV300, TT Electronics, peak wavelength $\lambda = 850\ \text{nm}$), which illuminated the coverslip from above and created inline holograms of the collected aerosols. These digital holograms were recorded as time-lapsed images at 2 fps by a complementary metal–oxide–semiconductor (CMOS) image sensor chip (Sony IMX219PQ, pixel pitch $1.12\ \mu\text{m}$) below the impactor, while aerosols were being sampled. Time-lapsed inline holograms of the captured particles were recorded at 2 fps. After the air sampling was complete ($\sim 180\ \text{s}$), additional 360 holographic frames were acquired at the same frame rate to sample the evaporation process of the captured volatile particles.

Hologram Reconstruction. The image sensor that was used to capture the holograms of e-cig-generated aerosols was a Bayer color

image sensor, which has slightly different responses for the four Bayer channels under the infrared illumination (850 nm). To correct for this difference, we performed a wavelet transform (using the fifth-order symlet) on each of the Bayer channels individually to estimate a low-frequency background shade image. Each channel was divided by their corresponding shade image to correct for the illumination shade and channel imbalances due to the Bayer filters. These corrected holograms were each digitally back-propagated by $750\ \mu\text{m}$ to roughly reach the top of the coverslip surface using the angular spectrum method.^{37,38} Zero padding was used in the angular spectrum domain to effectively up-sample the back-propagated image by two times in the spatial domain. The twin image noise and defocus artifacts in the back-propagated holograms were removed by a convolutional neural network (CNN)¹³ that was specifically trained on e-cig-generated aerosols to transform a randomly defocused, back-propagated hologram image to a phase-recovered, in-focus image, which achieves holographic phase recovery and autofocusing at the same time and generates an extended depth of field image reconstruction. The network structure was the same as in ref 13. To provide training data for this CNN, we used experimental data for the e-cig samples that were captured independent of the test results reported in this manuscript. Each hologram was split into smaller patches of 512×512 pixels. These patches were each autofocused and phase-recovered using a modified Gerchberg–Saxton error reduction-based phase recovery algorithm,^{39–41} which was only used during the CNN training phase. The original hologram patches were randomly back-propagated to an axial range of $\sim 200\ \mu\text{m}$ above and below the determined focal plane, which were used as input to the network during its training. The network was built on TensorFlow. The training took $\sim 30\ \text{h}$ for ~ 100 epochs on a laptop computer with a 3.60 GHz CPU, 16 GB RAM, and an Nvidia GeForce GTX 1080 GPU. After the training phase, which is a one-time effort, the trained CNN was blindly used to autofocus and phase-recover the images of the captured particulate matter. The network inference time is $<0.1\ \text{s}$ for a back-propagated hologram patch of 512×512 pixels and $\sim 10\ \text{s}$ for a full-FOV image of 3280×2464 pixels.

Aerosol Detection. A threshold level that equals to five standard deviations away from the image mean was applied to the real and imaginary channels of each reconstructed image and a spatial mask was generated as the union of the thresholded real and imaginary channels to mark the detected particle locations. Spatial masks for each time frame were stacked together, forming a space-time mask stack. Then, individual particles were detected by finding continuous traces within the space-time mask stacks. Empirically, we have seen that a volatile particle evaporates typically within 5–30 frames (2–15 s), as shown in, e.g., Supporting Information Movie S1, whereas a nonvolatile particle remains unchanged, in terms of volume and shape, during the entire imaging process. The acquired holographic image sequence of each detected particle (e.g., Figure 1d) was then used to determine the volatility per particle, covering a sample field-of-view of $4\ \text{mm}^2$; the details of this volatility measurement are presented in the next subsection.

Geometrical Parameter Fitting and Volatility Extraction.

The volatility of a sessile droplet landing on a substrate can be described by the decay rate of the contact angle (θ) between its edge and the substrate.^{11,42} Therefore, using these holographic images, the change in the contact angle of each particle with respect to the substrate was extracted as a function of time (t) using the reconstructed phase image channel. The collected volatile particles were assumed to be spherical caps positioned on the impactor surface. Given the phase value, $\text{ang}(s)$, of each pixel s , particle volume (V_i) was defined as an optical phase integral inside the identified mask area Ω_i of a given image frame i and was calculated using the illumination wavelength (λ) and a refractive index difference of $\Delta n = 0.4$ with respect to air, which is a typical value for PG and VG,^{43,44} i.e.,

$$V_i = \frac{\lambda}{2\pi\Delta n} \int_{\Omega_i} \text{ang}(s) \, ds \quad (1)$$

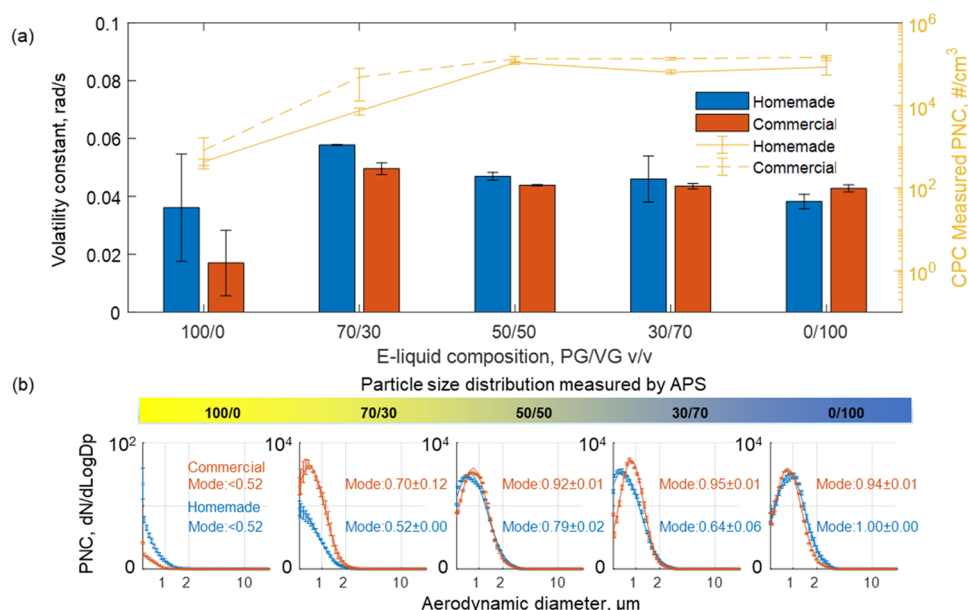


Figure 2. Volatility measurements and particle size distribution of particles generated by nicotine-free e-liquids with different PG/VG compositions (PG/VG ratio = 100/0, 70/30, 50/50, 30/70, and 0/100). (a) Volatility measurement results of different e-liquids by c-Air and particle number concentration (PNC) measurements by CPC. (b) Particle size distributions measured by APS. A log-normal distribution was fitted to each particle size distribution to calculate the mode diameter. The histograms of the measured volatility constants and size distributions can be found in Supporting Information Figure S1.

The maximum height h_i for each particle was calculated using the maximum phase value inside the mask, i.e.,

$$h_i = \frac{\lambda}{2\pi\Delta n} \cdot \max(\text{ang}(s)) \quad (2)$$

The contact angle for each individual particle (defining a spherical cap with h_i maximum height on the substrate) was calculated as

$$\theta_i = \arccos \left(1 - \frac{3h_i^2}{\frac{3V_i}{\pi h_i} + h_i^2} \right) \quad (3)$$

The contact angle decay rate K_i (rad/s) of each detected particle i was fitted using a linear model, i.e.,

$$\theta_i(t) = \theta_{i0} - K_i \cdot t \quad (4)$$

For each e-liquid sample, three independent measurements were conducted and n different particles (typically $n > 10\,000$ per measurement) were collected and analyzed. Measured angle decay rates for each particle ($K_1, \dots, K_n$) were fit to a Gaussian distribution, and the mean value of the fitted Gaussian distribution was treated as the calculated volatility constant K across all of the measured particles.

RESULTS AND DISCUSSION

Real-Time Imaging and Quantification of e-cig-Generated Particles for Different e-Liquid Compositions. PM volatility statistics and size distribution measurements for homemade and commercial e-liquids with varying PG/VG ratios are reported in Figure 2 and Supporting Information Table S1. As the PG/VG ratio of homemade (or commercial) e-liquid decreased from 70/30 to 0/100, the measured volatility constant K decreased from 0.058 ± 0.0002 rad/s (0.050 ± 0.002 rad/s) to 0.038 ± 0.0023 rad/s (0.043 ± 0.001 rad/s). For the homemade e-liquids, we found that the volatility constant is negatively correlated with the volumetric ratio of VG within the e-liquid ($p < 0.05$) except for the case of PG/VG = 100/0, suggesting that e-cig-generated particles become less volatile as the proportion of VG increases in the e-

liquid. This might be due to the fact that VG has substantially lower vapor pressure than PG (i.e., $P_{\text{sat(VG)}} \approx 20$ Pa while $P_{\text{sat(PG)}} \approx 0.1$ Pa at standard conditions), and this may lower the volatility of e-cig aerosols as the PG/VG ratio is reduced, in accordance with the Raoult's law. The negative correlation is consistent with a previous study that estimated the effects of PG/VG ratios on e-cig aerosols' particle loss rates due to evaporation, surface deposition, coagulation, and gravitation settling.³⁰ Our results further confirm the importance of evaporation in e-cig particle loss mechanisms and the effects of e-liquid components on the e-cig particles' volatility.

It is also noteworthy to mention that the volatility of aerosols produced from pure PG may be considerably underestimated in our measurements at 2 fps because of its fast-evaporating nature. As required by the sampling protocol of this study, the generated particles underwent a mixing and dilution process in the test chamber for ~ 26 –30 s before they are sampled by c-Air (see the Materials and Methods section). As a result, pure-PG particles that were captured and processed by c-Air were substantially lower in count. The PNC generated from pure PG was 1–2 orders of magnitude lower than other combinations of PG and VG, under the same particle generation protocol, i.e., a single puff of the e-cig (see Figure 2a). In accordance with our observations, PG-generated aerosols were previously reported to have higher particle loss rates due to their fast-evaporating nature compared to other e-liquid compositions.^{30,32}

c-Air volatility measurements can also be compared against the PNC and particle size distributions that were simultaneously recorded for each sample (see Figure 2a,b). As the PG/VG ratio shifted from 100/0 to 50/50, PNC inside the test chamber increased by more than 2 order of magnitudes (e.g., for homemade liquids from 435 ± 124 to $1.09 \pm 0.09 \times 10^5$ #/cm³). Further increase of VG concentration did not lead to a major PNC increase. The rapid PNC increase at lower VG levels may indicate that mostly the VG content in the e-liquid

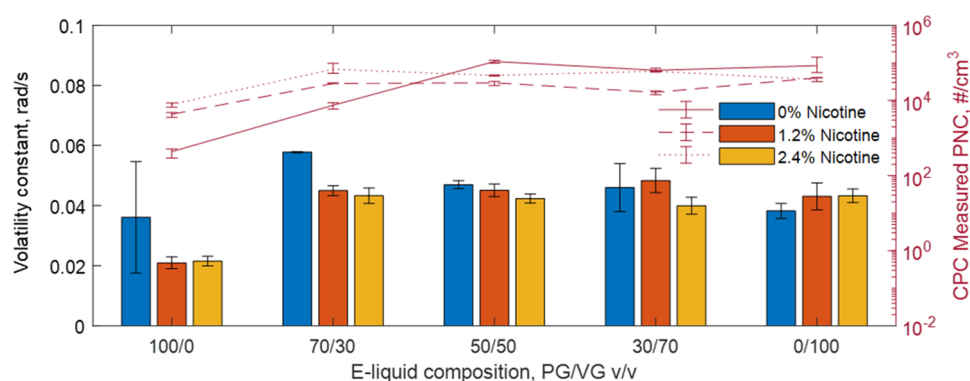


Figure 3. Volatility measurements of particles generated by puffing homemade e-liquids with different PG/VG compositions (PG/VG ratio = 100/0, 70/30, 50/50, 30/70, and 0/100) and nicotine levels (0, 1.2, and 2.4%). Volatility histograms are shown in Supporting Information Figure S2.

contributed to the aerosols that were generated. The particle size distributions in Figure 2b report the particle size (in μm) in x -axis and the normalized PNC (dN/dLogDp) in y -axis. As the PG/VG ratio in the e-liquid decreased, the mode of particle size distribution shifted from <0.5 to ~ 1.0 μm . The pure-PG particles in the e-cig vapor with their smaller size likely have higher volatility, as smaller liquid particles with greater vapor pressure evaporate faster due to the Kelvin effect.⁴⁵

Overall, our experimental results and analyses indicate that e-cig-generated particles evaporate slower with decreasing PG/VG ratios, along with a reduction in volatility with increasing particle size.

Impact of Nicotine on the Evaporation Behavior of e-cig-Generated Aerosols. While the solvents PG and VG are used to provide a tobacco-like smoking experience, e-cigs are also used as electronic nicotine delivery devices with varying levels of nicotine.⁴⁶ The effect of nicotine level on particle volatility is a key aspect in understanding e-cig aerosol dynamics. We examined the impact of three different nicotine levels (i.e., 0, 1.2, and 2.4% in mass concentration) on the volatility of e-cig aerosols generated by five different combinations of PG/VG ratios, as shown in Figure 3. The addition of nicotine significantly reduced the volatility of particles with high PG composition ($p < 0.05$). The measured volatility for pure-PG particles (and 70/30 PG/VG particles) reduced from 0.036 ± 0.019 rad/s (0.058 ± 0.001 rad/s) to 0.022 ± 0.002 rad/s (0.043 ± 0.003 rad/s), when 1.2% of nicotine was added into a nicotine-free e-liquid. The volatility across different PG/VG ratios remained at a similar level for 1.2%-nicotine-added e-liquids with a PG/VG ratio that is smaller than 70/30. Moreover, increasing the nicotine concentration from 1.2 to 2.4% did not further reduce the volatility for different PG/VG compositions.

These observations are also supported by the PNC and particle size distribution measurements (Supporting Information Figure S2). For pure-PG particles, adding 1.2% nicotine increased the PNC by 1 order of magnitude, from $4.35 \times 10^2 \pm 1.24 \times 10^2$ to $4.29 \times 10^3 \pm 6.63 \times 10^2$ $\#/\text{cm}^3$. Furthermore, adding 1.2% nicotine increased the PNC produced by 70/30 PG/VG e-liquid to the same order of magnitude as those with higher VG compositions generated. These results are consistent with prior studies^{47,48} that also reported an increase in PNC of e-cig aerosols as a result of the addition of nicotine. Similar to c-Air measurement results, no statistically significant change in PNC was observed between 1.2 and 2.4% nicotine-added e-liquid samples. Interestingly, the decrease in the

volatility that we observed in PG/VG mixtures with increasing VG levels was no longer evident after the nicotine was introduced into the e-liquid. In a simple two-component system, the PG/VG ratio determined the saturation vapor pressure of the e-liquid mixture, which dominated the volatility behavior of e-cig-generated aerosols. However, the addition of nicotine fundamentally alters the evaporation process, forming a more complex mixture with a different volatility behavior as we observed; these observations are also in good agreement with previous results.³⁰

Impact of Flavoring Additives on the Volatility of e-cig-Generated Particles. In addition to nicotine levels, we also evaluated how flavoring additives changed the evaporation dynamics of e-cig-generated particles. Both commercially available flavored e-liquids (e.g., tobacco, menthol, and strawberry flavors) and flavorless (no flavoring additives added) e-liquids were tested, with the flavorless e-liquid serving as the baseline for volatility comparisons. To avoid confounding factors from different PG/VG ratios and nicotine levels, we used 50/50 PG/VG nicotine-free e-liquids for all four tests. Our results, summarized in Figure 4, indicate that flavoring additives significantly changed the volatility of e-cig-generated aerosols. The volatility constant of flavorless e-liquid-generated aerosols was measured to be 0.044 ± 0.0002

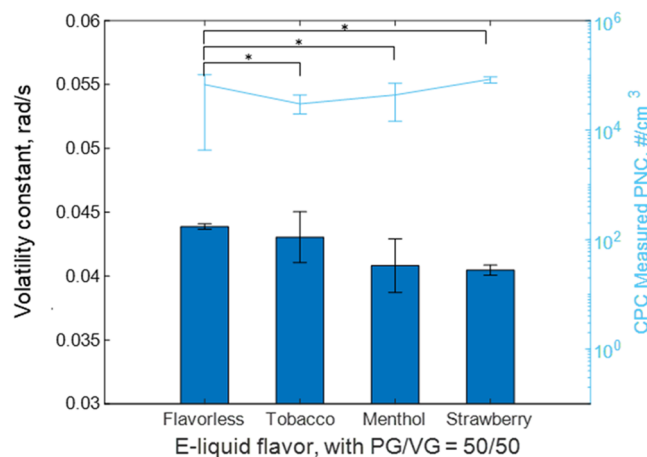


Figure 4. Volatility measurements of particles generated by puffing nicotine-free commercial e-liquids with different flavors (flavorless, tobacco, menthol, and strawberry flavor, with PG/VG = 50/50). Paired t -test $*p < 0.05$. Volatility histograms are shown in Supporting Information Figure S3.

rad/s, while the volatility constants of three flavored e-liquids were measured as 0.043 ± 0.002 rad/s for tobacco flavor, 0.041 ± 0.002 rad/s for menthol flavor, and 0.041 ± 0.0004 rad/s for strawberry flavor. A paired t-test was performed to compare the distribution of volatility constants between the flavorless e-liquid and each one of the flavored e-liquids, respectively, where a statistically significant change was obtained for all of the flavors (each with $p < 0.05$). Overall, these measurements reveal that the addition of these flavoring compounds into the e-liquid reduced the volatility of e-cig-generated aerosols.

Although a statistically significant change in volatility was observed in our c-Air measurements, the impacts of e-liquid flavoring on the particle size distribution and PNC measurements were minimal (Figure 4). When compared with the strong shifts observed in the particle size distributions due to the varying PG/VG ratios shown in Figure 2, the subtle changes introduced by flavoring additives indicate that the particle generation process is mainly governed by the PG/VG ratio (in the absence of nicotine). Nevertheless, the unknown and complicated chemical compositions and concentrations of these flavoring additives still complicate the evaporation behavior of e-cig-generated particles, as shown in our volatility measurements.

Future Work and Conclusions. In this work, we studied the evaporation dynamics of PM generated by e-cigs and we believe that understanding these dynamics might open up new avenues for e-cig-related research. Benefiting from its dynamic measurement capability, c-Air platform can also be used in the studies that examine second-hand vaping aerosols, where the aerosol volatility could heavily affect their behavior in the environment, resulting in different exposure mechanisms for public.^{14,49} The presented method and measurement device can also serve as a powerful tool in investigating particle/gas partitioning of any atmospheric aerosols, in addition to e-cig-generated aerosols. Therefore, the methodology used in this work can be applied to various fields that require dynamic measurements of volatile particles using portable devices.

In conclusion, we examined the volatility of e-cig-generated aerosols using lensless microscopy and deep learning. A negative correlation between e-cig-generated particle volatility and VG concentration in the e-liquid was revealed. The measured volatility constant K decreased from 0.058 ± 0.0002 to 0.038 ± 0.0023 rad/s, while the PG/VG ratio of a homemade e-liquid decreased from 70/30 to 0/100. The addition of other chemicals (e.g., nicotine and flavoring compounds) reduced the overall volatility of e-cig-generated aerosols ($p < 0.05$). The results obtained with the high-throughput c-Air device are consistent with previous studies that used traditional measurement methods.^{30,31} The presented approach can help us better examine the dynamic behavior of e-cig aerosols in a high-throughput manner, potentially providing important information for e-cig exposure assessment via, e.g., second-hand vaping.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssensors.1c00628>.

Histograms of volatility and particle size distributions of aerosols generated by nicotine-free e-liquids (Figure S1), by e-liquids with different nicotine levels (Figure S2), and by nicotine-free e-liquids with different flavors

(Figure S3); volatility comparison between emissions from a tank-type e-cig and a pod-type e-cig (Figure S4); table of volatility, PNC, and mode of particle size distributions of aerosols generated by nicotine-free e-liquids (Table S1) (PDF)

Movie of the evaporation process of a detected e-cig-generated aerosol (Movie S1) (MP4)

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Author Contributions

Y.L., Y.W., and L.L. contributed equally to this work. Y.L., Y.W., and E.C. contributed to the algorithms. Y.L., Y.W., L.L., Y.G., E.C., and A.O. contributed to the analyses. Y.L., Y.W., L.L., Y.G., and E.C. performed the experiments. All authors contributed the manuscript editing. A.O. and Y.Z. supervised the research. A.O. initiated the project.

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Notes

The authors declare no competing financial interest. The deep learning models reported in this work used standard libraries and scripts that are publicly available in TensorFlow. All of the data and methods needed to evaluate the conclusions of this work are present in the main text and Supporting Information. Additional data can be requested from the corresponding author (A.O.).

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