Formation of Oxidized Gases and Secondary Organic Aerosol from a Commercial Oxidant-Generating Electronic Air Cleaner

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ABSTRACT: The COVID-19 pandemic increased the demand for indoor air cleaners. While some commercial electronic air cleaners can be effective in reducing primary pollutants and inactivating bioaerosol, studies on the formation of secondary products from oxidation chemistry during their use are limited. Here, we measured oxygenated volatile organic compounds (OVOCs) and the chemical composition of particles generated from a hydroxyl radical generator in an office. During operation, enhancements in OVOCs, especially low-molecular-weight organic acids, were detected. Rapid increases in particle number and mass concentrations were observed, corresponding to the formation of highly oxidized secondary organic aerosol (SOA) (O:C ~ 1.3), with an enhanced signal at m/z 44 (CO2+) in the organic mass spectra. These results suggest that organic acids generated during VOC oxidation contributed to particle nucleation and SOA formation. Nitrate, sulfate, and chloride also increased during the oxidation without a corresponding increase in ammonium, suggesting organic nitrate, organic sulfate, and organic chloride formation. As secondary species are reported to have detrimental health effects, further studies should not be limited to the inactivation of bioaerosol or reduction of particular VOCs, but should also evaluate potential OVOCs and SOA formation from electronic air cleaners in different indoor environments.

INTRODUCTION

Many people spend most of their time indoors, making air quality in these spaces an important factor for human health. Indoor air quality (IAQ) depends on several factors, including but not limited to exchange with outdoor air, filtration, emissions from indoor sources, chemical reactions, e.g., via oxidation or multiphase processes, and deposition onto surfaces. Due to the impact of IAQ on health, there is a growing demand for air cleaning technologies meant to reduce exposure to potentially detrimental substances indoors. This demand has increased considerably during the course of the recent COVID-19 (severe acute respiratory syndrome coronavirus 2, SARS-CoV-2) pandemic due to the increased recognition of the role of airborne virus transmission, especially indoors.

Air cleaners are usually deployed with the intention to remove indoor pollutants such as particles or volatile organic compounds (VOCs) as well as to inactivate pathogens. Two types of air cleaning technologies are commonly used to remove particles: mechanical filtration and electronic air cleaners (e.g., ionizers and electrostatic precipitators). Gaseous pollutants such as VOCs and odoriferous compounds can be removed via a number of different technologies: adsorbent media air filters (e.g., activated carbon) and various electronic air cleaning devices that generate ions, reactive species, or other chemical products such as photocatalytic oxidation (PCO), plasma, and ozone-generating equipment, among others. In addition, hydroxyl radical (OH) generation via photolysis of ozone or water is also used to destroy odors and VOCs, usually as a substitute for ozone-generating air cleaners. Generally, it has been shown that ultraviolet germicidal irradiation (UVGI), ionizers, ozone generators, and PCO purifiers can be capable of inactivating viruses, bacteria, and other bioaerosol. However, their efficacy in real environments is disputed since their effectiveness can vary depending on the operating conditions (e.g., bacteria can repair damage if dosed with insufficient UV radiation) and there are no standardized testing conditions.

There are increasing concerns regarding the use of electronic air cleaners, as these devices can potentially generate unintended byproducts via oxidation chemistry similar to that in the atmosphere. The oxidation mechanism of VOCs in the atmosphere can be simplified as the following: (1) initial attack of the VOCs by oxidants (OH, O3, and NO3),...
(2) organic peroxy radical reactions, and in some cases (3) alkoxy radical reactions. Organic peroxy radicals can react with other species in the atmosphere (e.g., NO, NO₂, HO₂, etc.) and undergo functionalization or form alkoxy radicals. Alkoxy radicals can fragment and form smaller organic compounds in the atmosphere that can be oxidized further. Fragmentation leads to increased volatility whereas functionalization decreases volatility and increases solubility in water. These complex, multigenerational, gas-phase oxidation processes result in the formation of a large variety of organic compounds, which can undergo gas-particle partitioning and/or nucleation to form secondary organic aerosol (SOA). Such atmospheric processing was reported indoors during air cleaner operation. Previous studies reported secondary pollutant formation from ionizers, PCO systems with UV lamps, electrostatic precipitators, and plasma systems. These technologies were found to generate gas-phase byproducts such as ozone and less-oxidized VOCs (e.g., aldehydes) as well as secondary particles (e.g., ozone reaction with terpenes from air fresheners). However, there are no studies that report more-oxidized VOCs (e.g., carboxylic acids) or aerosol composition, which require advanced instrumentation.

In this work, we evaluated the effect of a commercial electronic air cleaner (hydroxyl radical generator) operated inside an office. We monitored gas-phase oxidized products and PM₁ (particulate matter less than 1 µm in diameter) size distribution and composition. We show that the operation of this device leads to the formation of small organic acids and increases PM₁ number and mass concentrations. As some byproducts of VOC oxidation can have adverse health effects, these results show that care must be taken when choosing an adequate and appropriate air cleaning technology for a particular environment and task.

### Materials and Methods

The experiment was performed in an office (∼16 m²), which was unoccupied except for instrument setup, in the Ford Environmental Sciences and Technology Building at the Georgia Institute of Technology. We performed the experiment in the following sequence: (1) 2.33 h of office background sampling, (2) 1.5 h of hydroxyl generator operation (Titan model #4000, International Ozone Technologies Group, Inc., Delray Beach, FL), and (3) 1.5 h of sampling after the device was turned off. Briefly, the device generates OH radicals and hydrogen peroxide (H₂O₂) via photocatalytic reaction of TiO₂ with UV-A range (365 nm) light and H₂O and O₂ in the air. Many other brands of hydroxyl generator are available in the market and employ a similar technology.

Gas-phase organic compounds were measured using a high-resolution time-of-flight chemical ionization mass spectrometer (HR-ToF-CIMS, Aerodyne Research Inc., Billerica, MA) with iodide (I⁻) as a reagent ion, which selectively measures some types of oxygenated organics. The instrument was not calibrated to report the mass concentrations of detected species and the signals were reported in counts per second. O₃ and NO₂ were monitored using an O₃ Analyzer (T400, Teledyne, City of Industry, CA), a NO-NO₂-NOₓ Analyzer (42C, Thermo Fisher Scientific, Waltham, MA), and a Cavity Attenuated Phase Shift NO₂ monitor (CAPS, Aerodyne Research Inc.).

Size-resolved PM₁ number and volume concentrations were measured using a scanning mobility particle sizer (SMPS, 17 nm–1 µm). The SMPS is a combination of a differential mobility analyzer (DMA) (TSI 3040, TSI Inc., Shoreview, MN) and a condensation particle counter (CPC) (TSI 3775). In addition, we deployed a separate CPC (TSI 3025 A) to monitor the total number concentration of particles (all particles under roughly 3 µm). Chemical composition of particles smaller than 1 µm was monitored using a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, Aerodyne Research Inc.). HR-ToF-AMS quantifies nonrefractory species (organics, nitrate, sulfate, ammonium, and chloride) mass concentrations and measures the bulk elemental composition of the particles (e.g., O:C and H:C ratios) via particle vaporization on heated surface (∼600 °C) and electron ionization (70 eV). The elemental ratios for particles were calculated based on the “Improved-Ambient” method to correct for molecular functionality-dependent biases which can originate from the vaporization and ion fragmentation processes in the instrument.

### Results

**Formation of Oxygenated VOCs (OVOCs).** The immediate formation of oxygenated products was observed by the HR-ToF-CIMS (Figure 1) when the device was turned on. Formic acid (m/z 173, CH₃O₂⁻), nitrous acid (m/z 174, HONO¹⁻), acetic acid (m/z 187, C₂H₄O₂⁻), iminoacetic acid (m/z 200, C₂H₄NO₂⁻), oxamide (m/z 215, C₃H₆N₂O₂⁻), glyceraldehyde (m/z 217, C₃H₅O₂⁻), glycerol (m/z 219, C₃H₆O₄⁻), alanine (m/z 216, C₃H₆NO₂⁻), and acetoacetic acid (m/z 229, C₄H₈O₄⁻) were identified and showed the most obvious enhancements during the operation period. Enhanced glyceraldehyde and glycerol at the beginning of the experiment (12:10 pm) was likely due to the presence of people in the office initially (to set up instruments for this study), as these compounds are formed as intermediates in metabolism and widely used in cosmetics or as an additive in foods.

In contrast, glycolic acid (m/z 217, C₃H₄O₃⁻), succinic acid (m/z 245, C₄H₈O₄⁻), hydrogen peroxide (m/z 161, H₂O₂⁻), nitric acid (m/z 190, HNO₃⁻), and dinitrogen pentoxide (m/z 235, N₂O₅⁻) decreased during the operation period. While glycolic acid and succinic acid decreased continuously throughout the experiment, they decreased faster during the device operation. These two compounds are widely used in skin care products and are applied in various commercial goods (e.g., food, pharmaceuticals, polymers, paint, cosmetics), respectively.

Hydrogen peroxide increased during the background period and decreased during the operation of device. As mentioned in the previous section, TiO₂ photocatalytic technology is reported to produce H₂O₂ as another product. However, H₂O₂ decreased when the device was turned on and rebounded after the device was turned off. The pre-existing H₂O₂ in the office could have been reacting with the generated OH radicals but flattened as a result of regeneration via self-reaction of hydroperoxyl radicals. The increase in HONO and decrease in HNO₃ and N₂O₅ during the device operation could have been a consequence of increasing particle surface area during the device operation via new particle formation (Figure 2). Surface reactions indoors have been reported to play a role as a source of HONO but as a sink for HNO₃ and N₂O₅.

**Formation of Secondary Organic Aerosol.** Particle number and volume concentrations started increasing once the device was in operation (Figure 2a). Both the number and volume concentrations increased rapidly in the first 30 min...
after the device was turned on and slowed down after reaching \(~4000\) particles cm\(^{-3}\) and \(~5\) \(\mu\)m\(^3\) cm\(^{-3}\), respectively. This was followed by a rapid decrease in concentrations after the device was turned off. The increase in particles was mostly within the PM\(_1\) size range based on the agreement between the SMPS (PM\(_1\) only) and the CPC (all particles under roughly 3 \(\mu\)m). During the operation, an enhancement was observed in the 100–200 nm size range for both particle number and volume concentrations (Figure S2). It is noted that a similar experiment was performed in a laboratory space (\(~140\) m\(^2\)) and similar results were observed (Figure S3).

The time series of the species measured by the HR-ToF-AMS are shown in Figure 2b. A collection efficiency of 0.45 was applied to the data as the inorganic concentrations were low and the aerosol did not contain high mass fractions of acidic sulfate or ammonium nitrate.\(^5\)\(^2\) The chemical composition of the particles during the operation period confirmed SOA formation, with organics reaching 2.1 \(\mu\)g m\(^{-3}\) after the device was turned on. Figure 2 also shows that the mass concentrations of nonrefractory species reported by the HR-ToF-AMS were somewhat lower than the volume concentration enhancement measured by the SMPS (converted to mass concentration by the density of each species; Figure S4). This difference was expected since both instruments sampled particles without the use of a dryer at the instrument inlets. Thus, the particle concentration reported by the SMPS included water whereas particle water can be evaporated in the low-pressure aerodynamic lens and vacuum system of HR-ToF-AMS.\(^5\)\(^2\)\(^5\)\(^5\)\(^5\) The discrepancy diminishes when accounting for particle water as shown in Figure S4. Ammonium concentration was low throughout the experiment and showed little changes whereas nitrate, sulfate, and chloride increased during the device operation period. Since we did not observe ammonium increasing along with nitrate, sulfate, or chloride, these species are likely in the form of organic nitrate, organic sulfate, and organic chloride. Organic mass spectra shows enhanced fraction at \(m/z\) 44 (CO\(_2^+\)) during the device operation (Figures 3a and S5), with increased O:C and decreased H:C (Figures 3b and S6a). Note that organic contribution to \(m/z\) 28 (CO\(^+\)) is set to be equal to the value at \(m/z\) 44 during data analysis,\(^4\)\(^3\)\(^5\)\(^6\) hence the signals at these two ions are the same. The increase in the degree of oxidation of aerosol is further illustrated in the Van Krevelen diagram in Figure 3b.\(^5\)\(^7\)\(^5\)\(^9\) The aerosol evolution followed a slope of \(~-1\), with particle carbon oxidation state (OS\(_C\) = 2 O:C - H:C)
increasing during device operation, as a result of enhancements in O:C and reductions in H:C.60

**DISCUSSION**

Generation of hydroxyl radicals indoors reduces VOC concentrations in a similar manner to tropospheric VOC oxidation chemistry, which proceeds through complex, multi-generational chemistry and results in the formation of a large number of organic products. The byproducts formed from these reactions in this study depend on the identity of the VOCs in the office. VOCs were not measured in this work; however, VOC concentrations in multiple different office buildings in the United States were reported to be dominated by acetone, ethanol, 2-propanol, toluene, xylene, limonene, dichloromethane, 1,1,1-trichloroethane, 2-propanone, n-pentane, n-tetradecane, n-pentadecane, and n-hexadecane.61−64 It is possible that the office in this study has a similar VOC speciation, though it could have a lower total carbon budget due to no occupancy and no activities, such as printing,65 which can lead to VOC emissions. The observed oxygenated C3−C6 compounds can be formed from functionalization and fragmentation of such VOCs during the oxidation.24,32 Although we only observed small carboxylic acids in the gas phase, this does not exclude the formation of aldehydes and larger OVOCs,32 which might not be detected by I-CIMS, be lost in the instrument inlet lines, interact with surfaces,66 or participate in new particle formation and growth.67 Increasing particle number and mass concentrations and the formation of highly oxidized SOA suggest that new particle formation and condensation growth can be a loss process of larger, less-volatile OVOCs. The SOA formed has an O:C of ∼1.3, which is higher than the typical O:C range observed for more-oxidized oxygenated organic aerosol (MO-OOA) in ambient environments.58,68 The observed nucleation is likely due to the small condensation sink with low aerosol background (∼581 particles cm−3) in the office. The enhancement of m/z 44 (CO2+) in the HR-ToF-AMS organic mass spectra indicates the contribution of organic acids in SOA formation, as their thermal decarboxylation gives rise to the CO2+ fragment.58,69−72 In the Van Krevelen diagram, the SOA evolved along the ∼−1 line, which corresponds to the addition of carboxylic acids and/or simultaneous increases in alcohol and carbonyl groups.73,74 Taken together, these results show that carboxylic acids were formed during the oxidation process and contributed to new particle formation owing to their low volatility.24,75 The enhancements in particle-phase carboxylic acids measured by the HR-ToF-AMS are in agreement with the formation of gas-phase carboxylic acids measured by the I-CIMS.

Nitrate, sulfate, and chloride enhancements are expected to be associated with organic nitrate, organic sulfate, and organic chloride formation (Figure S6). The average NO+/NO2+ ion ratio from the HR-ToF-AMS is widely used as an indicator to differentiate inorganic vs organic nitrate.74−76 The NO+/NO2+ ratio for inorganic nitrate during the instrument calibration was 1.98, and previous laboratory studies have shown that this ratio is much higher for organic nitrate than inorganic nitrate.74,77−79 The average NO+/NO2+ ratio during the operation period was ∼17, implying that virtually all the particle-phase nitrate was organic nitrate. The contribution of organic sulfate can be examined by evaluating the fractions of HSO3− and H2SO4− in HSO4− fragments (SO−, SO2−, SO3−, HSO3−, SO2−, and H2SO4−).80 Both fractions decreased when the device was turned on, implying the presence of organic sulfate. Organic chloride formation can be associated with chlorine-containing VOCs which may be emitted or formed through interactions with cleaning products.81,82

To our knowledge, this is the first study that monitored the chemical composition of secondary products in both gas and particle phases during the operation of an electronic air cleaner that dissipates oxidants in a real-world setting. Although we lack parent VOC measurements (I-CIMS has low sensitivity toward less- or nonoxidized VOCs), limited number of OVOCs, small enhancements in SOA, and negligible changes in ozone and NOx (Figure S7) observed during this work were assumed to be due to low initial VOC concentrations in the office where this study was conducted. Much larger enhancements in OVOCs, SOA, and ozone could be observed in other types of indoor environments such as industrial settings, homes, and restaurants, which can have much larger VOC concentrations, even more than in outdoor locations.1,66,82−84 Secondary VOC oxidation products have been shown to have detrimental effects on human health.33,35,36,45,58,85 Specifically, SOA has been reported to induce cellular reactive oxygen species (ROS) generation, inflammatory cytokine production, and oxidative modification of RNA.87−89 The toxicity of SOA...
could increase with increasing $\text{OS}_\text{c,}^{17,90}$ Therefore, future studies on air cleaning technologies should not be limited to the inactivation of bioaerosol or reduction of particular VOCs, but should also evaluate potential OVOCs and SOA formation during their operation. Further studies need to be conducted to comprehensively investigate how different factors and conditions impact secondary chemistry, including testing in different indoor environments with various device settings as well as evaluating uniformity across a larger number of devices and reproducibility in measurements. The electronic air cleaner tested in this study is similar to many other commercially available devices, and similar experiments should be conducted with other devices.

### ASSOCIATED CONTENT

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.1c00416.

HR-ToF-CIMS mass spectra, size-dependent particle number and volume and concentrations, SMPS and CPC results from an experiment conducted in a larger space (laboratory) than the office, AMS and SMPS total mass comparison and AMS particle water correction, AMS organic families mass spectra, time series of AMS $\text{O}_\text{c}, \text{H}_\text{c}$, and $\text{NO}_\text{c}/\text{NO}_\text{c}^+$ ratios, and time series of ozone, NO, $\text{NO}_2$ (PDF)

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**Notes**

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