Ethanol-based disinfectant sprays drive rapid changes in the chemical composition of indoor air in residential buildings

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ABSTRACT

The COVID-19 pandemic has resulted in increased usage of ethanol-based disinfectants for surface inactivation of SARS-CoV-2 in buildings. Emissions of volatile organic compounds (VOCs) and particles from ethanol-based disinfectant sprays were characterized in real-time (1 Hz) via a proton transfer reaction time-of-flight mass spectrometer (PTR-TOF-MS) and a high-resolution electrical low-pressure impactor (HR-ELPI+), respectively. Ethanol-based disinfectants drove sudden changes in the chemical composition of indoor air. VOC and particle concentrations increased immediately after application of the disinfectants, remained elevated during surface contact time, and gradually decreased after wiping. The disinfectants produced a broad spectrum of VOCs with mixing ratios spanning the sub-ppb to ppm range. Ethanol was the dominant VOC emitted by mass, with concentrations exceeding 10^3 mg m^{-3} and emission factors ranging from 10^1 to 10^2 mg g^{-1}. Listed and unlisted diols, monoterpenes, and monoterpenoids were also abundant. The pressurized sprays released significant quantities of nano-sized particles smaller than 100 nm, resulting in large deposited doses in the tracheobronchial and pulmonary regions of the respiratory system. Inhalation exposure to VOCs varied with time during the building disinfection events. Much of the VOC inhalation intake (>60%) occurred after the disinfectant was sprayed and wiped off the surface. Routine building disinfection with ethanol-based sprays during the COVID-19 pandemic may present a human health risk given the elevated production of volatile chemicals and nano-sized particles.

1. Introduction

The COVID-19 pandemic has led to the surging use of disinfectants for inactivation of SARS-CoV-2 (U.S. CDC, 2020; Pitol and Julian, 2021; Zheng et al., 2020). Ethanol (C2H5OH) can inactivate human coronaviruses, including SARS-CoV-2 (Kratzel et al., 2020; Kumar et al., 2021; Meyers et al., 2021; Rabenau et al., 2005). Ethanol-based disinfectants have been widely used during the COVID-19 pandemic and are included in the U.S. EPA List N: Disinfectants for Coronavirus (U.S. EPA, 2020). Ethanol-based disinfection of buildings may pose a health risk due to inhalation exposure to ethanol and other volatile organic compounds (VOCs) included in the disinfectants.

Inhalation exposure to gas-phase ethanol is associated with adverse toxicological outcomes. Preclinical studies on animals revealed potential neural, hepatic, pulmonary, and cardiovascular risks of inhalation of ethanol and 2-propanol (Budygin et al., 2007; Hirth et al., 2016; Mouton et al., 2016). An increase of multiple biomarkers (BrAC, EtG, and EtS) was observed after use of alcohol-based hand sanitizers (1–3 pumps every 2–20 min) in several studies (Ahmed-Lecheheb et al., 2012; Ali et al., 2013; Arndt et al., 2014; Hautemanière et al., 2013a, 2013b). Inhalation of ethanol is of concern as it can be directly transmitted to the brain via arterial circulation, which may increase the risk of addiction (Maclean et al., 2017).

Given the diverse spectrum of ingredients included in commercial ethanol-based disinfectants, it is expected that such products can emit a wide range of VOCs. The human respiratory system, as the main route of...
VOC intake (Gong et al., 2017; Zhao et al., 2019), is at risk of impairment to pulmonary function when exposed to elevated VOC concentrations (Yoon et al., 2010; Zhao et al., 2019). Epidemiological studies have identified associations between inhalation exposure to VOCs and numerous respiratory diseases (Elliott et al., 2006; He et al., 2015). VOC exposures in residential and office buildings have been related to various adverse human health outcomes (Cakmak et al., 2014; Wolkoff et al., 2006). Pressurized sprays can also release sub-10 μm droplets and sub-100 nm particles; the composition of which is influenced by the spray product applied (Bekker et al., 2014; Hagendorfer et al., 2010; Jiang et al., 2021a; Nazarenko et al., 2011). Sub-100 nm particles can penetrate deep into the human respiratory system and result in adverse health outcomes, including respiratory and cardiovascular diseases (Heinzerling et al., 2016; Knibbs et al., 2011; Li et al., 2016; Oberdörster, 2000; Oberdörster et al., 2005; Stone et al., 2017; Weichenhal et al., 2007; Downward et al., 2018; Fanas et al., 2014; Soppa et al., 2014).

Despite the potential exposure risks associated with increased use of disinfectant sprays during the COVID-19 pandemic, there is limited research characterizing the temporal emission profiles of such products. The objective of this study is to conduct real-time (1 Hz) emission measurements of gas- and particle-phase species released from ethanol-based disinfectants through use of a proton transfer reaction time-of-flight mass spectrometer (PTR-TOF-MS) and a high-resolution electrical low-pressure impactor (HR-ELPI+), respectively. Such state-of-the-art instrumentation enables improved temporal characterization of the fate and transport of potentially health hazardous materials in the indoor environment during building disinfection activities and associated inhalation exposures among occupants. To the authors’ knowledge, this represents the first tandem application of a PTR-TOF-MS and HR-ELPI+ for real-time detection of VOCs and particles released from ethanol-based disinfectants.

### 2. Materials and methods

#### 2.1. Study site: Purdue zero Energy Design Guidance for Engineers (zEDGE) Tiny House

The measurement campaign was conducted in a residential architectural engineering laboratory – the Purdue zero Energy Design Guidance for Engineers (zEDGE) Tiny House (Fig. S1). zEDGE is a mechanically ventilated single zone residential building with a conditioned interior volume of 60.35 m³. A powered ventilator with a MERV 16 filter supplied filtered outdoor air to zEDGE (Fig. S2). zEDGE was maintained at a positive pressure (+9 Pa) with an outdoor air exchange rate (AER) of 3 h⁻¹. The details of the AER calculation are provided in the supplementary material. Two fans were used to promote mixing.

#### 2.2. Protocol for ethanol-based disinfectant emission experiments

The disinfectant emission experiments were conducted on two 2 ft. × 3 ft. glass panels positioned on the kitchen countertop of zEDGE. The glass panels were used to isolate the indoor surface onto which the disinfectant was applied. A clean set of glass panels was used for each experiment. Two ethanol-based disinfectant sprays included in List N disinfectant was applied. A clean set of glass panels was used for each glass panels in a matrix with 3 rows and 8 columns at about 1 ft. above the surface. At the end of the second period, the liquid film of disinfectant was removed via wiping with two absorbent towels (removed from zEDGE after wiping). The third period included a 60 min concentration decay. zEDGE was occupied by one researcher during the spray and wipe events and unoccupied for the remaining periods. The disinfectants were used as prescribed by the manufacturer.

##### 2.3. Real-time measurement of VOCs via PTR-TOF-MS and particles via HR-ELPI+

Mixing ratios of VOCs were measured at 1 Hz by a PTR-TOF-MS (PTR-TOF 4000, Ionicon Analytic Ges.m.b.h, Innsbruck, Austria) using hydronium (H₃O⁺) as the reagent ion. In the drift tube, molecules with proton affinities greater than that of water (691 kJ mol⁻¹) will collide with H₃O⁺ and be ionized through a proton transfer reaction (Blake et al., 2009; Lindinger et al., 1998). The ionized molecules are then detected by a TOF-MS. Mass spectra for mass-to-charge ratios (m/z) from 20 to 450 were recorded. Pressure, voltage, and temperature for the drift tube were set at 2.2 mbar, 600 V, and 70 °C respectively, maintaining the ionization field energy (E/N) at approximately 139 Td. The abundance of impurity ions (O₆⁺, NO⁻) was < 10 %. The sampling inlet was located near the center of the zEDGE kitchen. A PFA tube (3/8 in. OD) was used as the sampling line. At the intake of the sampling line, a PTFE membrane filter (1 μm pore size) was installed to remove particles. The exhaust from the PTR-TOF-MS was directed outdoors. The PTR-TOF-MS was calibrated daily using two VOC gas standard mixtures (Table S2). Mixing ratios of VOCs not included in the gas standards were calculated based on proton transfer reaction theory (de Gouw and Warneke, 2007; Klein et al., 2018). Reaction rate constants were derived from the literature if available (Pagonis et al., 2019), otherwise 2 × 10⁻9 cm³ s⁻¹ was used (Table S3). Given the large number of ions detected by the PTR-TOF-MS during the disinfection events, only the ions that increased by > 50 % beyond their average background mixing ratios and peaked at > 0.1 ppb are reported. A material balance model was used to estimate ethanol emission factors (EFs, mg g⁻¹ or mg ml⁻¹), as described in the supplementary material.

Particle size distributions from 6 to 10000 nm in aerodynamic diameter (Dₐ) were measured at 1 Hz with a HR-ELPI+: (Dekati Ltd., Kangasala, Finland). The aerodynamic diameter of an irregularly shaped particle is the diameter of a spherical particle with a density of 1 g cm⁻³ and the same settling velocity as the irregularly shaped particle (Hinds, 1999). The HR-ELPI+ sampled air near the center of the zEDGE kitchen. Oil-soaked sintered collection plates were used to eliminate particle bounce and impactor overloading. Details on the operational principle of the ELPI+ can be found elsewhere (Järvinen et al., 2014; Lemmetty et al., 2005; Marjamäki et al., 2009; Marjamäki et al., 2005). The HR-ELPI+ uses an iterative inversion algorithm to improve the size-resolution beyond the standard ELPI+: (Saari et al., 2018). The exhaust of the HR-ELPI+ pump was directed outdoors via the bathroom exhaust (Fig. S2). Measured particle number size distributions (dn/dLog Dₐ) were translated to particle mass size distributions (dm/dLog Dₐ) assuming spherical particles with a density of 1 g cm⁻³ for Dₐ<100 nm (Wu and Boor, 2020) and the density of the disinfectant solution for particles with Dₐ>100 nm, which were likely generated through the spray process (Table S1). The particle number size distributions were used to estimate number respiratory tract deposited doses for the head airways, tracheobronchial region, and pulmonary region, as described in the supplementary material.
3. Results and discussion

3.1. Temporal VOC emission profiles during ethanol-based disinfection events in buildings

The use of ethanol-based disinfectants in buildings results in sudden changes in the chemical composition of indoor air. Products A and B emitted a complex mixture of VOCs. More than 50 ions were found to increase by >50% relative to background, with individual ion mixing ratios spanning the sub-ppb to ppm range (Tables S4–S5). Detected ions were assigned to one of the following VOC categories: (1.) ethanol and ethanol cluster ions; (2.) listed fragrance compounds for product B; and (3.) compounds associated with essential oils. Remaining ions with identified ion formulas were categorized as: (4.) hydrocarbons (C_xH_y); (5.) oxygenated hydrocarbons (C_xH_yO_z); and (6.) nitriles (C_xH_yN) for product B. Signals without identified formulas were reported as: (7.) unidentified. Experiments A2 and B1 were selected as case studies for each product. The real-time PTR-TOF-MS data was visualized in two forms: time-series of ion-resolved mixing ratios (Fig. 1 (I.a., II.a.)) and time-series of categorized VOC mass concentrations (Fig. 2). Figs. S4–S11 show results for other experiments.

Several of the detected ions are associated with ethanol. m/z 47.06 (C_2H_5O^+) is the parent ion of ethanol, m/z 45.03 (C_2H_5O^-), m/z 65.06 (C_2H_5O^-(H_2O)), m/z 75.08 (C_2H_5^+(C_2H_5O)), m/z 93.10 (C_2H_5O^-(C_2H_5O)), and m/z 139.12 (C_2H_5O^-(C_2H_5O)_2) are fragment or cluster ions of ethanol (Boscaini et al., 2004; Buhr et al., 2002). The formation of ethanol cluster ions in the drift tube of the PTR-TOF-MS at high ethanol mixing ratios, such as those observed here, is described in detail by Boscaini et al. (2004). The increase of m/z 31.02 (CH_3O^+) may be due to reactions between ethanol and O_2^- impurity ions in the drift tube (Kushch et al., 2008; Schripp et al., 2010; Spanel and Smith, 2008).

Ethanol can react with the hydroxyl radical and nitric oxide to form formaldehyde (CH_2O) and acetaldehyde (C_2H_4O), which are detected at m/z 31.02 and m/z 45.03, respectively (Dunmore et al., 2016). However, the PTR-TOF-MS cannot separate isomers and fragment ions of different origins; thus, the potential indoor formation of formaldehyde and acetaldehyde cannot be evaluated. Real-time separation of isomers requires a PTR-TOF-MS configured with a fast gas chromatograph.

Fig. 1. (I.) Time-series for ethanol-based disinfection event A2 using product A. (a.) ion-resolved mixing ratios from m/z 31 to 193, (b.) particle number size distributions from D_a = 6 to 300 nm, and (c.) particle mass size distributions from D_a = 300 to 10000 nm. (II.) Time-series for ethanol-based disinfection event B1 using product B. (a.) ion-resolved mixing ratios from m/z 31 to 247, (b.) particle number size distributions from D_a = 6 to 300 nm, and (c.) particle mass size distributions from D_a = 300 to 10000 nm. Additional details on the detected ions can be found in Table S3. Peak and average background mixing ratios for each ion can be found in Tables S4–S5. Peak and average background particle number and mass concentrations can be found in Table S6.
During experiment A2, the ethanol mass concentration increased immediately after the disinfectant was applied, reaching a peak at 14.64 mg m$^{-3}$ (6574 ppb), 200-fold greater than background levels (0.073 mg m$^{-3}$, Table S4) (Figs. 1(a.) and 2(a.)). Ethanol concentrations remained elevated during the 10 min surface contact time, after which they gradually decayed following removal of the disinfectant residue from the glass panels. However, the ethanol mass concentration remained at 0.62 mg m$^{-3}$ (326 ppb) at the end of the 60 min decay period, 9-fold greater than background. A similar temporal emission profile was observed during experiment B1, where the ethanol mass concentration peaked at 18.97 mg m$^{-3}$ (7983 ppb), 600-fold greater than background (0.034 mg m$^{-3}$, Table S5) (Figs. 1(b.a.) and 2(b.)). Ethanol was the dominant VOC released from both disinfectant sprays by mass (Fig. 2). This is due in part to the abundance of ethanol in the disinfectant solutions (A: 30–60 %wt., B: 58 %wt.) and the high volatility of ethanol (Salthammer, 2016). The mean ethanol EFs for disinfectants A and B were 99.32 mg g$^{-1}$ (82.54 mg ml$^{-1}$) and 32.31 mg g$^{-1}$ (26.17 mg ml$^{-1}$), respectively (Table S7). This suggests that disinfection activities are important episodic indoor sources of ethanol.

In addition to ethanol, the PTR-TOF-MS measurements revealed the presence of a myriad of VOCs in the disinfectant emissions; their temporal emission profiles can be seen in Figs. 1 and 2. While product A did not specify fragrance ingredients, monoterpenes and monoterpenoids related to essential oils were detected by the PTR-TOF-MS (Angulo Milhem et al., 2020; Nematzolahi et al., 2018); this enables characterization of the unlisted fragrances in the disinfectant. Mixing ratios of monoterpenes (C$_{10}$H$_{16}$, detected at m/z 81.07 (C$_{6}$H$_{10}$) and m/z 137.14 (C$_{10}$H$_{17}$)) peaked at 10.7 ppb after applying product A. Monoterpenoids detected at m/z 151.13 (C$_{10}$H$_{15}$O$^+$, possibly thymol or carvone), m/z 153.12 (C$_{10}$H$_{14}$O$^+$, possibly camphor or citral), m/z 155.13 (C$_{10}$H$_{15}$O$^+$, possibly linalool, eucalyptol, citronellal, a-terpineol, or terpinen-4-ol), and m/z 193.16 (C$_{13}$H$_{27}$O$^+$, possibly ionone or damascone) are components of essential oils (Babu et al., 2002; Kokkini et al., 1995; Msaada et al., 2007; Wang et al., 2009). Their mixing ratios increased 2.7-4.2-fold above background, but were relatively low in magnitude, with peaks ranging from 0.33-0.76 ppb. Many of the listed fragrance components for product B (>0.01 %wt. or on a designated list) were detected by the PTR-TOF-MS. m/z 135.09 (C$_{6}$H$_{13}$O$^+$, dipropylene glycol), m/z 137.14 (C$_{8}$H$_{17^+}$, 3-carene, also detected at m/z 81.07), m/z 157.15 (C$_{10}$H$_{15}$O$^+$, dihydromyrcenol), m/z 165.09 (C$_{10}$H$_{13}$O$^+$, eugenol), m/z 197.17 (C$_{10}$H$_{23}$O$^+$, linalyl acetate), and m/z 247.21 (C$_{17}$H$_{27}$O$^+$, acetyl cedrene) reached peak mixing ratios 2.6-18.2-fold greater than background. Several unlisted monoterpenoids were also observed (m/z 151.13, m/z 153.12, m/z 155.13, and m/z 193.16). Their mixing ratios were 4.1-12.1-fold greater than background, however, peak mixing ratios were low (1.0-1.6 ppb). Monoterpenes and monoterpenoids are common fragrance ingredients in disinfectants and could cause occupational asthma (Malo and Chan-Yeung, 2009; Melchior Gerster et al., 2014; Vizcaya Fernández et al., 2011).

Emissions from the two ethanol-based disinfectants led to significant increases in the mixing ratios (typ. 100–450 ppb) of m/z 42.04 (C$_{3}$H$_{4}$), m/z 43.05 (C$_{3}$H$_{7}$), and m/z 57.07 (C$_{3}$H$_{8}$). These ions can be attributed to the high abundance of alkanes (often ≥5 %wt.) included in the sprays as propellants (Dinh et al., 2015). C$_{3}$H$_{4}^+$, C$_{3}$H$_{7}^+$, and C$_{3}$H$_{8}^+$ can be formed due to reactions between i-C$_{3}$H$_{10}$ and n-C$_{3}$H$_{10}$ with O$^+$ impurity ions (Spanel and Smith, 1998; Wilson et al., 2003). Similarly, C$_{3}$H$_{4}$ can be formed via reactions between C$_{3}$H$_{8}$ and O$^+$ and NO$^+$ impurity ions. While i-C$_{3}$H$_{10}$ and n-C$_{3}$H$_{10}$ have low proton affinities (677.8 kJ mol$^{-1}$), it has been shown they undergo slow association reactions with H$_{2}$O$^+$ to form C$_{3}$H$_{4}$H$_{2}$O$^+$ (Wilson et al., 2003). Due to such reactions with H$_{2}$O$^+$ reagent ions and O$^+$ and NO$^+$ impurity ions, PTR-TOF-MS measurements in the indoor environment are therefore sensitive to episodic emissions of C$_{3}$-C$_{4}$ alkanes from pressurized spray-based consumer products. This precludes identification of selected ingredients in the disinfectants, such as triethylene glycol and t-butanol, that have a major fragment at m/z 57.07 (Pagonis et al., 2019). Several unlisted diols were also detected by the PTR-TOF-MS, including methanediol (m/z 49.03), 1,2-ethanediol...
(m/z 63.04), 1,2-propanediol (m/z 77.06), 1,4-butanediol (m/z 91.07), and diethylene glycol (m/z 107.08). Peak mixing ratios for most were between 1–10 ppb, with methanediol the most abundant at 18–28 ppb.

3.2. Temporal particle emission profiles during ethanol-based disinfection events in buildings

Application of ethanol-based disinfectant sprays was associated with a significant increase in indoor particle number (PN) and particle mass (PM) concentrations (Table S6). The temporal evolution in particle number and mass size distributions during disinfection events A2 and B1 are illustrated in Fig. 1(b., II.b.) and (I.c., II.c.). The HR-ELPI+ measurements revealed substantial formation of nano-sized particles with \( D_a \leq 100 \) nm (modal \( D_a \approx 23 \) nm) immediately after application of product A. Peak PN concentrations ranged from 8.3 to 11.5 \( \times 10^4 \) cm\(^{-3} \), similar in magnitude to PN concentrations observed during cooking on gas and electric stoves (Jiang et al., 2021b). Product A also produced accumulation and coarse mode particles from \( D_a = 300–10000 \) nm that dominated particle mass size distributions. PM\(_{2.5}\) and PM\(_{10}\) concentrations built up rapidly to 706 \( \mu g \) m\(^{-3}\) and 2708 \( \mu g \) m\(^{-3}\), respectively. Product B produced fewer particles compared to A. The variation in particle emissions among the two products may be due to differences in the spray nozzles (Nuyttens et al., 2007) and the composition of the disinfectant solutions (Kim et al., 2020). The observed particles are likely disinfectant droplets formed by the propellant spray process (Bekker et al., 2014; Hagendorfer et al., 2010).

Indoor production of nano-sized particles on the order of \( 10^4-10^5 \) cm\(^{-3} \) by the ethanol-based disinfectant sprays resulted in number respiratory tract deposited doses of \( 10^8-10^9 \) particles for an adult during the spray and contact periods (Table S8). Much of the number dose was received in the tracheobronchial and pulmonary regions. Such sprays may present a health risk as inhalation of sub-100 nm particles is associated with numerous adverse toxicological outcomes (Allen et al., 2017; Hussein et al., 2020; Li et al., 2016; Stone et al., 2017).

3.3. Human inhalation exposure to VOCs during ethanol-based disinfection events in buildings

High-resolution VOC measurements with a PTR-TOF-MS enables evaluation of real-time human inhalation exposure to VOC emissions during and after application of ethanol-based disinfectants. Inhalation exposure was evaluated for an adult involved in an indoor disinfection event following the temporal sequence of the experiments: 2 min spray period, 8 min contact period, 2 min wipe period, and 58 min decay period. Fig. 3 illustrates the per-period inhalation intake of VOCs for events A2 and B1. Figs. S12–S15 show results for other experiments. Ethanol contributed 80 % and 83 % to the total VOC inhalation intake during disinfection events A2 and B1, respectively. A non-negligible fraction (>15 %) of the total VOC inhalation intake was attributed to the remaining VOC categories (Fig. 3). Thus, one will inhale a complex mixture of VOCs during application of ethanol-based disinfectants. The magnitude of the total VOC mass inhaled (7–8 mg) presents a health risk for those involved in routine building disinfection during the COVID-19 pandemic.

The PTR-TOF-MS revealed time-dependent variations in VOC inhalation intake during the disinfection events. The spray, contact, wipe, and decay periods contributed 3 %, 24 %, 6 %, and 67 % to the total VOC inhalation intake (7.54 mg) during the 70 min disinfection event A2,
The study demonstrated that a PTR-TOF-MS and HR-ELPI+ can be used together for near-real-time indoor detection of gas- and particle-phase species during residential building disinfection activities. Ethanol-based disinfectant sprays emitted a complex multi-phase mixture that included a broad spectrum of VOCs and particles. Notably, the disinfectants released significant quantities of ethanol (6000–8000 ppb) and nano-sized particles (10^4–10^5 cm^-3) to the indoor environment. Inhalation exposure was strongly time-dependent, with the majority of VOC inhalation intake occurring in the one hour period after the disinfectant was sprayed and wiped off the surface. The increased use of ethanol-based disinfectants during the COVID-19 pandemic, there is an urgent need to understand the health risks of exposure to elevated concentrations of VOCs and particles produced by such products. As shown in this study, PTR-TOF-MS and HR-ELPI+ measurements can characterize the emissions of potentially health hazardous materials released from disinfectant sprays.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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Appendix A. Supplementary material

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.haz.2021.100042.

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