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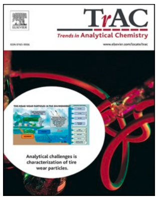

Tracking indoor volatile organic compounds with online mass spectrometry

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ABSTRACT

Humans spend most of their time indoors, where they are exposed to many volatile organic compounds (VOCs). Exposure to indoor VOCs has been linked to adverse health effects, and so a comprehensive indoor air quality assessment requires a strong fundamental knowledge of indoor VOCs. The recently developed advanced online mass spectrometry enables fast, non-targeted measurements for VOCs, which provides a powerful tool to track the complex indoor VOCs. This review article summarizes the principles of different online mass spectrometry techniques and their application to indoor VOC measurements. The sources, emission characteristics, and chemical compositions of primary indoor VOCs are discussed. Recent advances in the fundamental understanding of chemical transformations and formation mechanisms related to secondary indoor VOCs are also discussed. By summarizing the current advances and remaining challenges for the investigation of indoor VOCs, we propose future research directions regarding their origin, transformation chemistry, environmental fate, and health impact.

1. Introduction

Humans spend more than 80 % of their time indoors, where they are exposed to numerous structurally diverse gaseous organic compounds [1,2]. These gaseous organic compounds can be divided into four main categories according to their saturation vapor pressures (C^*): volatile organic compounds (VOCs, $C^* > 3 \times 10^6$ μg m^{-3}), intermediate-volatility organic compounds (IVOCs; 300 μg $m^{-3} < C^* < 3 \times 10^6$ μg m^{-3}), semi-volatile organic compounds (SVOCs; 0.3 μg $m^{-3} < C^* < 300$ μg m^{-3}), and low-volatility organic compounds (LVOCs; $C^* < 0.3$ μg m^{-3}) [3,4]. Indoor VOCs and I/S/LVOCs can enter human body through inhalation and air-to-skin dermal uptake. Human exposure to indoor VOCs has been linked to many adverse health effects such as respiratory and cardiovascular diseases [5–7]. To mitigate the potential health risks induced by these harmful chemicals, in 2010, the World Health Organization (WHO) has set up indoor air quality management guideline for certain VOCs such as benzene and formaldehyde [8]. Therefore, understanding the chemical compositions (i.e., what are they?), sources (i.e., where do they come from?), emission rates (i.e., what are the concentrations indoors?), and chemical transformations of indoor VOCs (i.e., will they transform to other compounds with different physicochemical properties and different toxicity?), is of great importance for the corresponding health risk evaluation.

Compared to outdoor environments, indoor environments have

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several distinguishing features. First, the surface-area-to-volume ratio (S/V) indoors is typically around 3 m^{-1} (estimated based upon available macroscopic surfaces such as furnishings and walls), which is orders of magnitude higher than the S/V outdoors [9]. This value likely represents a lower limit to the actual S/V at the microscopic scale, considering that many indoor surfaces (e.g., bedding and carpets) have porous structures which contribute to a higher effective surface area [10]. This makes many VOCs in the outdoor air behave as SVOCs indoors [11]. Second, the oxidant concentrations and light levels indoors are very different from outdoors. Due to the low light intensity indoors relative to outdoors, the levels of oxidants (e.g., OH radicals and O_3) are generally much lower in indoor environments [12]. Third, humans (e.g., human breath and human skin) and human activities (e.g., cooking and cleaning) have strong impacts on indoor VOCs [13]. As a result, VOCs indoors possess different emission sources and different transformation chemistry compared to VOCs outdoors.

Much attention has been paid to indoor VOCs since the 1970s, owing to its important role in determining indoor air quality [14–17]. Traditionally, indoor VOCs are measured with offline-targeted chemical analysis approach. In this approach, indoor VOCs and I/S/LVOCs are usually collected using passive samplers, canisters or sorbent traps, then analyzed with offline techniques such as gas chromatography-mass spectrometry (GC-MS) [18–20]. One of the limitations of this approach is that the time response is relatively slow (several hours or more), making it difficult to capture fast physical and chemical processes related to indoor VOCs. Recently, with the development of advanced online mass spectrometry such as proton-transfer-reaction time-of-flight mass spectrometry (PTR-ToF-MS) and chemical ionization mass spectrometry (CIMS), the indoor VOCs "measurement paradigm" has shifted from offline to online monitoring [21–23]. The use of these high resolution online mass spectrometry allows time-resolved (seconds to minutes), non-targeted measurements for indoor VOCs, thus providing a powerful tool to illustrate the "indoor chemical cocktail".

In this article, we summarize recent advances in studies relevant to indoor VOCs, with a focus on: (1) the principles, advantages, and limitations of online mass spectrometry for tracking indoor VOCs (Section 2), (2) sources and characteristics of primary indoor VOCs (Section 3), (3) characteristics and formation mechanisms of secondary indoor VOCs (Section 4). In Sections 3 and 4, we briefly mention early studies on this topic while mainly discussing the recent advances that have occurred in the past five years. Based on the current understanding of the sources, emissions, and transformation chemistry of indoor VOCs, we further provide recommendations for future research directions in Section 5. This article is distinguished from past reviews on this topic [15,16,24] by mainly focusing on new findings of indoor VOCs from both real-time laboratory studies and indoor measurements conduced over the past five years, and by providing a molecular-based insight.

2. Online mass spectrometry

The operation details of online mass spectrometry, including PTR-ToF-MS, Vocus PTR-ToF-MS (Vocus is a name of a new chemical ionization source that can be used for PTR-ToF-MS measurements), CIMS, semi-volatile thermal desorption aerosol gas chromatography (SV-TAG), and secondary electrospray ionization-mass spectrometry (SESI-MS), have been described in literature [25–29]. The principles of these instruments are briefly summarized below.

2.1. PTR-ToF-MS

PTR-ToF-MS is a soft ionization mass spectrometry technology which ionizes the analytes through proton transfer reactions: $H_3O^+ + M \rightarrow MH^+ + H_2O$; M stands for VOC molecules with proton affinity higher than H_2O (691 kJ mol^{-1}) [25]. The product ions MH^+ are detected by the ToF-MS. PTR-ToF-MS can measure VOCs at low level (pptv) with high-time resolution (seconds), and has become one of the most frequently used online mass spectrometry for indoor VOCs measurements.

For VOCs whose authentic standards are available, their sensitivities can be quantified with calibration experiments, i.e., introducing VOCs with known concentrations into a PTR-ToF-MS. For VOCs whose authentic standards are not available, their sensitivities can be predicted using the method developed by Sekimoto et al. [30]. Briefly, for a given VOC, its proton-transfer reaction rate coefficient ($k_{\text{proton-transfer}}$) can be estimated based upon its polarizability and dipole moment. With the estimated $k_{\text{proton-transfer}}$, the sensitivity can be obtained.

2.2. Vocus PTR-ToF-MS

Vocus PTR-ToF-MS is a recently developed instrument [26]. Compared to conventional PTR instruments, it has several new features. First, the new design of a discharge reagent-ion source and a focusing ion−molecule reactor (FIMR) improves the detection efficiency of product ions. The FIMR consists of a glass tube with a resistive coating on the inside surface and four quadrupole rods mounted radially on the outside. With a radio frequency field, ions are collimated to the central axis, thus minimizing their losses in the FIMR and leading to an enhanced detection efficiency. Second, due to the high water mixing ratio in the FIMR, the Vocus PTR-ToF-MS does not exhibit sensitivity dependence on the relative humidity (RH) of sample air, which is in contrast with the behavior of conventional PTR instruments for some species [31]. Consequently, Vocus PTR-ToF-MS has an enhanced sensitivity for VOCs (compared to conventional PTR-ToF-MS), and is able to measure I/SVOCs as well [32].

2.3. CIMS

Chemical ionization is a widely-used ionization method, which uses different types of reagents to chemically ionize molecules with various properties. Traditionally, quadruple CIMS is commonly used for target detection of a limited number of ions, given the unit mass resolution of the detector. With the development of mass spectrometry technique, chemical ionization in combination with ToF-MS is able to measure a wide variety of species in real-time. The most commonly used reagent ions include acetate, methyl iodide, nitrate, etc. for negative ionization [27,33,34] and ammonia, benzene, water, etc. for positive ionization [35–37]. For example, the acetate CIMS has good selectivity and sensitivity for inorganic and organic acids [11,34,38], while the nitrate CIMS has been used for measurements of oxidized organics in both field studies and laboratory experiments [32,39]. In the case of iodide CIMS, it can measure halogenated species, oxidized organics and nitrogen-containing inorganic species, etc. [27,40,41].

2.4. SV-TAG

SV-TAG is a GC-MS based technique that can measure SVOCs in both gas- and particle-phase simultaneously [42]. Briefly, a small flow of sample air passes through a $PM_{2.5}$ cyclone and then directs onto two parallel metal fiber filter sampling cells, enabling the collection of both gaseous and particle-bound SVOCs [43]. In comparison to the traditional offline methods that analyze SVOCs upon sample collection and extraction, SV-TAG has the advantage of high time-resolution and limited sample preparation. The simultaneous detection of both gas- and particle-phase SVOCs enables the investigation of gas-particle partitioning behaviors and surface emissions of SVOCs indoors [43–46].

2.5. SESI-MS

SESI-MS is a novel technique for real-time measurements of VOCs. VOCs in the sampling air are ionized upon interacting with electrospray droplets, and then detected by a high resolution mass spectrometer [47].

Spray electrification is achieved by inserting an electrified platinum wire, held at a high voltage (e.g., 2.5 kV), in the electrospray working solution [48]. The mass spectra can be recorded in positive or negative ion mode, with a time resolution of 1 s. The limits of detection for VOCs range from pptv to ppbv [49], depending on their structures. One of the limitations of SESI-MS is that the ionization efficiencies of VOCs vary from compound to compound, making it difficult to quantify the concentration of unknown chemicals (i.e., the chemicals whose authentic standards are unavailable). In this case, one may determine the concentration of unknown chemicals using the semi-quantification approach by assuming their sensitivities are the same as a VOC with known sensitivity [50].

2.6. Sampling strategy and data analysis

Online mass spectrometry measures VOCs through direct air sampling, i.e., indoor air is introduced into the instrument without sample pre-treatment. This provides a powerful analytical method to track the complex indoor air composition, which is changing frequently due to the continuous indoor-outdoor air exchange and the impact of environmental factors (e.g., temperature, humidity, and human activities) [11]. Also, the "fast time-response" feature of online mass spectrometry enables a useful sampling strategy, i.e., to obtain information of spatial variability during indoor measurements. Specifically, this can be achieved by setting up several sampling lines to different locations (e.g., kitchen, bedroom, and outdoors) and automatically switching between them [11,21,51].

The measurement frequency of indoor VOCs can range from hours to days, depending on the research purposes. In general, the following information can be obtained from real-time indoor measurements: chemical composition of VOCs, VOC concentrations, VOC emission rates, and the dynamic change of VOCs during measurements. Combining this information with indoor conditions (e.g., temperature, humidity, and occupants), further analysis can be conducted to illustrate the emission mechanism, fate, and exposure risks of indoor VOCs.

2.7. Advantages and limitations

In general, conventional PTR-ToF-MS is good at VOCs measurements, while Vocus PTR-ToF-MS, CIMS, and SV-TAG are able to detect I/SVOCs [32,42]. SV-TAG has an hourly time resolution, which is longer than those of PTR-ToF-MS and CIMS (seconds). Therefore, researchers should consider these features when selecting online mass spectrometry to track indoor VOCs.

Compared to the offline chemical analysis approach, online mass spectrometry has several advantages. First, online mass spectrometry can capture the dynamic changes of indoor VOCs due to its high time resolution. This is particularly important for monitoring fast indoor processes such as the dynamic gas-surface partitioning process of indoor VOCs [11]. This is also important for the measurements of indoor VOCs with high reactivity (i.e., having short lifetimes). Offline collection and analysis may lead to sampling and measurement artifacts for these reactive species. Second, offline analysis typically requires complicated sample preparation procedures (sample collection, extraction, and analysis), while online mass spectrometry measures indoor VOCs through direct sampling (which is relatively simple). Third, the recently developed online mass spectrometry (e.g., Vocus PTR-ToF-MS) are highly sensitive, which can detect indoor VOCs at very low level (pptv). This makes it possess the ability to identify low-concentration emerging indoor VOCs such as the newly discovered liquid crystal monomers [52].

Despite these advantages, certain limitations exist for online mass spectrometry. First, these instruments are expensive, "noisy" and require large space during sampling, which may hinder their application in indoor measurements (particularly for the measurements in residences). Second, online mass spectrometry is unable to distinguish isomers, unless combining a GC with these instruments [53]. Third, the interpretation of the complex mass spectra is challenging given the potential fragmentation effect during measurements. It requires strong knowledge and skills to analysis the high-resolution mass spectra data.

3. Primary indoor VOCs

Primary indoor VOCs have been extensively studied over the past few decades. Early studies on this topic mainly focus on the VOCs emitted from indoor surfaces such as building materials, furniture, carpet, and paint. These static contents represent the conventional, well-known sources of primary indoor VOCs. Recently, increasing attention has been paid to primary VOCs related to humans, human activities, and microorganisms, as well as emerging VOCs from commercial products. Given this, we briefly mention early studies on primary VOCs in Section 3.1, while mainly discussing the recent advances that have occurred in the past five years in Section 3.2. The emission sources and characteristics of primary VOCs are summarized in Fig. 1 and Table 1.

3.1. Early studies of primary indoor VOCs

Measurement of primary indoor VOCs date back to the 1970s. In 1975, Andersen et al. measured the concentration of indoor formaldehyde released from chipboards (a common building material) in Danish dwellings, using the chromotropic acid method [14]. Since then, indoor air quality issues raised from static contents have attracted attention from researchers and the public. Research focus include investigating VOCs from different types of building materials (e.g., particle board, wall paper, and floor varnish), furniture (e.g., dining table, sofa, and desk chair), carpets, and paint [54–57]. During the 1980s and 1990s, studies on this topic were conducted with GC techniques (GC-MS and GC-flame ionization detection). While the reported emission factors of VOCs vary from one study to another (depending on the test materials and indoor conditions), a general conclusion is that these materials can emit a wide range of VOCs including alkanes, alkenes, terpenes, alcohols, aldehydes, ketones, esters, and aromatics, which have strong impact on indoor air quality [54–58].

The indoor environment features multiple dynamic processes that lead to rapid fluctuations of indoor air composition. Thus, direct monitoring of these processes is essential for understanding the underlying mechanism and for the determination of process-specific parameters. GC techniques are difficult to track the dynamic processes due to their slow time-response. With the development of proton-transfer-reaction quadruple mass spectrometry (PTR-QMS) in the late 1990s [59] and then PTR-ToF-MS (having higher mass resolution than PTR-QMS) in the early 2000s [60], the application of online mass spectrometry in indoor chemistry studies emerges [25]. Online mass spectrometry can capture fast indoor processes, which provides new data and new insights into the emission of indoor VOCs. This makes the research focus of indoor primary VOCs expanding from static contents to non-static sources, as discussed in Section 3.2.

3.2. Recent advances in primary indoor VOC measurements

3.2.1. Human-released VOCs

Human breath and human skin. Human breath and human skin serve as major sources for VOCs emitted from humans. Humans exhale many endogenous VOCs, which are generated during metabolic processes in human body. These exhaled compounds include acetone, isoprene, methanol, butyric acid, dimethyl sulfide, etc. [61]. The total emission factor of breath VOCs (EF_{breath}) can be determined using environmentally controlled chambers combined with online PTR-ToF-MS measurements [62]. Under O_3-free conditions, the estimated EF_{breath} is 1290 μg h^{-1} $person^{-1}$, with acetone, isoprene, and methanol being the three most abundant VOCs, which contribute 59 %, 19 %, and 13 % of the EF_{breath}, respectively [63].

Human skin-released VOCs are more diverse than VOCs in the breath

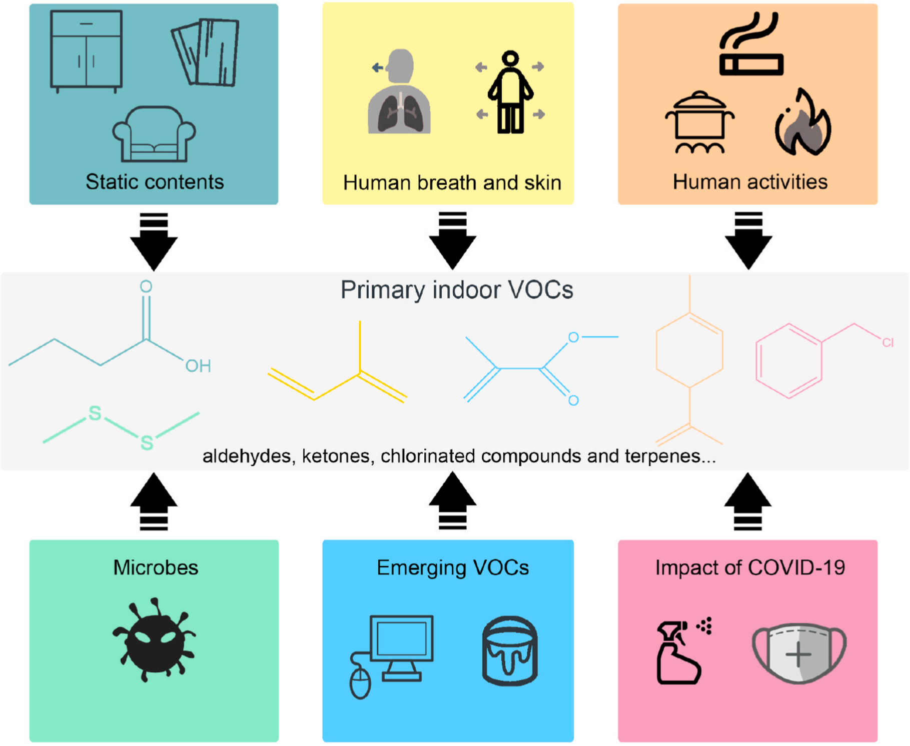

Fig. 1. Overview of the emission sources of primary indoor VOCs.

Table 1
Summary of primary indoor VOC emissions.

Sources		Instruments	Species	Emission factor/ concentration	Ref.
Human-released VOCs	Human breath	PTR-ToF-MS	Acetone, isoprene, methanol, etc.	1.29 × 10^3 μg person^{-1} h^{-1}[a]	[63]
	Human skin	PTR-ToF-MS	Acetic acid, acetone, acetaldehyde, etc.	1.15 × 10^3 μg person^{-1} h^{-1}[a]	[63]
Personal care products	Shamboo	CIMS	2-Propanol, benzyl alcohol, limonene, etc.	29.2 μg s^{-1} g^{-1}[a]	[70]
Emerging VOCs from commercial products	Computer monitor	Vocus PTR-ToF-MS	Acetic acid, isoprene, 1,9-nonanediol, etc.	5.19 × 10^9 molecules s^{-1} cm^{-2}[a]	[52]
	Laptop	Vocus PTR-ToF-MS	Acetic acid, cyclohexene, 1,9-nonanediol, etc.	8.25 × 10^9 molecules s^{-1} cm^{-2}[a]	[52]
	TV screen	Vocus PTR-ToF-MS	Acetic acid, 1,9-nonanediol, isoprene, etc.	2.90 × 10^9 molecules s^{-1} cm^{-2}[a]	[52]
	Coatings of furniture	PTR-QiToF-MS	Dibasic esters	4–41 μg m^{-3}	[94]
Cooking	Edible oils	PTR-ToF-MS	Acrolein, hexanal, heptanal, etc.	5–78 mg kg^{-1}[a]	[72]
	Peanut oils	SESI-HRMS	Decenoic acid, 9-oxononanoic acid, etc.	2.0–368.8[b]	[49]
	Fermented rice	PTR-ToF-MS	Cyclohexane, and ethanol	1.9[b]	[73]
	Amino acids and triglycerides	Vocus PTR-ToF-MS	$C_{2-11}H_{5-23}NO$, $C_{18}H_{35}NO$	33–813 μg g^{-1}[a]	[74]
	Making coffee and toasting bread	PTR-ToF-MS	Pyridine and ethanol	1.7–420 ppb	[21]
	Frying egg with toast	SV-TAG	Hexadecanoic acid and octadecanoic acid	/	[44]
	Oven-usage	SV-TAG	Squalene and low-volatility siloxanes (D13–D20 cyclic and L13-L19 linear siloxanes)	/	[45]
Smoking	Cigarette smoking	PTR-MS	2,5-Dimethylfuran, acetonitrile, cyclohexane, etc.	2.3–4.3[b]	[73]
	Thirdhand smoke	PTR-ToF-MS	Acetone, acetaldehyde, acetic acid, etc.	72.1 mg h^{-1}[a]	[80]
Burning	Solid fuel	PTR-ToF-MS	Acetic acid, acetaldehyde, phenol, etc.	6000 mg kg^{-1}[a]	[86]
	Candle	PTR-ToF-MS	Benzene	1.5[b]	[73]
	Mosquito-repellent incense	PTR-MS	Benzene	40.7–58.6 ppb	[88]
Impact of COVID-19	Natural product mopping	PTR-ToF-MS	Monoterpenes and citral	38 mg mopping^{-1}[a]	[22]
	Quaternary ammonium disinfectants	PTR-ToF-MS	Phenol, ethanolamine, phenoxyisopropanol, etc.	0.16–9.6 mg g^{-1}[a]	[96]
	Botanical disinfectants	PTR-ToF-MS	Thymol, $C_{10}H_{16}$, $C_{10}H_{14}O$, etc.	11.7–2701 μg g^{-1}	[98]
	Face masks	PTR-QiToF-MS	Methanol, acetonitrile, acetaldehyde	91–836 μg m^{-3}[a]	[108]

[a] Emission factors for total VOCs.
[b] The ratio of signals with and without indoor activities.

air. It was found that acetic acid and acetone were the two major compounds present in dermal VOCs, together accounting for 35 % of the total emission factor of dermal VOCs [63]. Other dermal VOCs include C_xH_y (hydrocarbons), and $C_xH_yO_{1\text{-}3}$ (aldehydes, ketones, and acids) [63, 64].

Air humidity, indoor temperature, and clothing type can affect human-released primary VOCs. Except for acetone and isoprene, the emissions of the other VOCs exhibited a positive relationship with humidity (32–62 % RH) and temperature (29–33 °C) [63]. Also, humans wearing short clothing were observed to emit more $C_xH_yO_2$ (e.g., acetic acid) than those wearing long clothing, because clean clothing can act as a physical barrier to hinder the release of VOCs from skin to the air [63]. Interestingly, a recent study found that the emission of isoprene from humans was strongly impacted by human emotional states during film watching (measured with a PTR-ToF-MS), suggesting that isoprene can be used as a chemical indicator for setting the age rating of films in the future [65].

Personal care products. Cyclic volatile methylsiloxanes (cVMS) are synthetical chemicals widely used as inactive ingredients in personal care products such as cosmetics, antiperspirants, and hair care products [66]. Octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) are the three most frequently detected cVMS in indoor air. Based on PTR-ToF-MS measurements in a university classroom, Tang et al. found that cyclic siloxane compounds were the most abundant VOCs emitted from humans, followed by acetone, and acetic acid [67]. Among the detected cVMS, D5 had the highest emission rate (up to 9800 μg h^{-1} $person^{-1}$), while the emission rates of D4 and D6 were one or two orders of magnitude lower than D5 [68,69]. The emission rate of D5 decreased from morning into the afternoon, which is consistent with human activity patterns, i.e., daily morning application of personal care products led to a higher emission rate of cVMS in the morning [68]. Considering this, D5 may be used as a tracer to track human-related VOCs indoors.

In addition to cVMS, personal care products can also emit other VOCs. Yeoman et al. detected high levels of limonene, methanol, and 2-propanol (along with D4 and D5) from different personal care products (shampoo, shower gel, moisturizer, and conditioner) used in the UK [70, 71]. The use of personal care products increases the probability for humans to inhale VOCs released from these products. It was estimated that the inhalation dose of limonene during the usage of facial moisturizer was 16 times higher than that inhaled from typical indoor ambient air over 24 h [71]. This highlights the need to comprehensively assess the potential health risk associated with personal care products-released VOCs.

3.2.2. Human activities

Cooking. Cooking plays an important role in determining indoor air quality, due to significant VOC emissions from cooking materials (e.g., oils, spices), stoves, appliances, etc. [21]. Oxygenated VOCs (OVOCs; mainly aldehydes and ketones) typically dominate VOC emissions from heating oils, but the dominant compounds and their EFs differ among cooking oils with different chemical components [72]. For example, oils rich in unsaturated fatty acids (oleic acid, linolenic acid, α-linolenic acid, etc.) are more prone to be oxidized during heating, forming VOC products such as nonanal acid, nonanoic acid, azelaic acid and 9-oxononanoic acid [73]. Heterocyclic compounds like 2-acetyl-5-methylfuran and 5-propyldihydro-2(3*H*)-furanone were detected with an SESI-HRMS during thermal processing of foods and were suggested as tracers for cooking emission [49]. When heating amino acids (representing protein-rich food materials) together with triglycerides, nitrogen-containing species were observed in both gas-phase ($C_{2\text{–}11}H_{5\text{–}23}NO$) and airborne particles (oleamide, $C_{18}H_{35}NO$) [74].

Cooking styles can also lead to different VOC emission behaviors. For instance, indoor pyridine and ethanol reached 1.7 ppb (background concentration 0.06 ppb) and 420 ppb (background concentration 80 ppb) in a California residence when making coffee and toasting bread, respectively [22]. Lunderberg et al. reported squalene and low-volatility siloxanes emissions associated with oven usage (measured with a SV-TAG) [45]. It is also known that nitrogen-containing VOCs (e.g., pyrrole, hydrogen cyanide, acetonitrile, etc.) were generated during usage of oven, but it remains an open question whether these VOCs were formed from heating nitrogen-containing food only or from the combination of food, coating and/or cleaner residues [75].

Smoking. Cigarette smoking emits large amounts of toxic gaseous pollutants indoors, leading to negative health effects on humans [76]. Nicotine and the related products (myosmine, isonicoteine and nicotyrine), other nitrogen-containing compounds (propanenitrile, pyridine, and pyrrole), BTEX (benzene, toluene, ethylbenzene, and xylenes), and alkenes are well-known VOCs from smoking [77,78]. Lyu et al. identified 2,5-dimethylfuran as a marker for smoking, with an emission ratio (ER, the ratio of signals with and without indoor activities) of 4.3 [73]. Alkanes and aldehydes which have high ERs are also notable fingerprints for smoking [73].

Thirdhand smoke (THS), the residue of tobacco smoke that is sorbed to indoor surfaces, has been suggested as an important exposure pathway to humans [79]. 2,5-Dimethylfuran, 2-methylfuran, and acetonitrile usually served as THS tracers [80]. Interestingly, these tobacco smoke markers have been detected in a strictly non-smoking theater, which likely arise from THS [80]. Note that surface-sorbed THS can partition to indoor aerosols and then enter human body through aerosol inhalation or dermal uptake [81,82].

As a “healthier and safer” alternative to tobacco cigarettes, electronic cigarettes (e-cigarettes), have attracted researchers’ attention recently due to their potential hazards to humans [83]. Breiev et al. demonstrated that a novel setup based on coupling heated inlet and PTR-ToF-MS measurements enables fast, high time resolution analysis of puff-by-puff variations in e-cigarette aerosols with online quantification [84]. Real-time measurements of VOC emissions from e-cigarette revealed that, in contrast to the conventional cigarettes, there was no significant puff dependence in the e-cigarette emissions [85]. In comparison to the traditional cigarette smoke, fewer studies focused on online measurements so far and further studies are needed to confirm the harmfulness of e-cigarettes.

Burning. In addition to smoking and cooking, burning activities including solid fuel, candles, mosquito-repellent incense, etc., can produce many gaseous compounds. For example, the EF of total VOCs was reported to be 6000 mg kg^{-1} when burning beech logs in a residential wood burner. Among them, acids (CH_2O_2 and $C_2H_4O_2$), carbonyls (C_2H_4O, C_3H_4O, and C_3H_6O), and aromatics (C_6H_6, C_7H_8, and C_6H_6O) were predominant species [86]. In the case of candle burning, hydrocarbons (C20–C40), long-chain fatty acids and their esters are main constituents of candle fuels, together with additive fragrances and colorants [87]. Notable increased levels of alkanes were observed when burning a ritual/scented candle in a kitchen. Furthermore, addition of fragrances results in more VOC emissions such as xylenes/ethylbenzene, ethyl acetoacetate, styrene, and crotonaldehyde [73]. With respect to mosquito-repellent incense burning, benzene accounted for the largest proportion of VOC emission from both the liquid mosquito-repellent and disc mosquito-repellent incense, as observed with a PTR-ToF-MS. Compared with liquid mosquito-repellent, disc mosquito-repellent incense burning leads to lower VOC EF, but higher particle emission; thus, more attention should be focused on its particle emission profiles [88].

3.2.3. Microbial VOCs

Microbial volatile organic compounds (MVOCs) are a variety of compounds from the metabolism of fungi and bacteria that grow on indoor surfaces and indoor dust, as well as from human activities [89, 90]. The concentrations of individual MVOCs range from ng m^{-3} up to mg m^{-3}, spanning several orders of magnitude [89]. Speciation of MVOCs is complicated, including oxygen-containing VOCs (e.g., small alcohols and carbonyls), terpenoid compounds, metabolites such as nitrogen- and sulfur-containing species, aromatics, etc. MVOCs also include species associated with odor, such as dimethyldisulfide, dimethyl sulfide, ethanethiol, methanethiol and methional [91,92]. The health effects of MVOC exposure relate to eye and upper-airway irritation, and allergenic responses, potentially leading to wheezing, asthma, and other lung malfunctions.

Most previous studies on MVOCs are based on offline sampling combined with GC-MS analysis. Below we summarize a few recent studies based on PTR-ToF-MS online measurements, which provides an opportunity to investigate MVOC emissions and the associated influencing factors in a time-resolved manner. The most critical factor affecting indoor microbial activities and their VOC emissions is moisture [93]. Haines et al. investigated microbial growth and the corresponding MVOC emissions from carpet, drywall and floor dust under different RH conditions [92]. They found microbial growth and MVOC emissions occurred at a relatively lower RH in carpet and floor dust (75 % RH) compared to drywall (85 % RH). Misztal et al. conducted a comprehensive characterization of the emission rates of MVOCs from environmental bacteria and fungi, and examined the factors that influence MVOC emissions [91]. The results suggest that in addition to moisture, the MVOC emission intensity and speciation are also affected by substrate type, microbial interaction structure, and life stage.

3.2.4. *Emerging VOCs from commercial products*

With the development of chemical industry, new commercial products which contain many synthetic chemicals are released into the market and are used indoors. These commercial products can serve as sources of emerging indoor VOCs. For example, a recent laboratory study found that liquid crystal displays (LCDs, including computer monitors, laptops, and TV screens) can emit more than 40 gaseous compounds, with a total screen area–normalized emission rate of up to 8.25×10^9 molecules s^{-1} cm^{-2} [52]. The detected gaseous compounds include 10 emerging VOCs (liquid crystal monomers; a key component used in liquid crystal displays), and more than 30 other VOCs (e.g., acetic acid, isoprene, and nonanal). In addition, based on PTR-ToF-MS measurements in new apartments in Beijing, China, another recent study found dibasic esters (a group of emerging VOC) were present in indoor air, with concentrations of up to 41 μg m^{-3} [94]. The newly identified dibasic ester contaminants were likely sourced from the coatings of furniture. From a human health perspective, emerging VOCs indoors may lead to new human exposure risks. Therefore, further study is needed to investigate the source, emission characteristics, and toxic effects of these emerging VOCs.

3.2.5. *Impact of COVID-19*

Disinfectants-related VOCs. The COVID-19 pandemic has resulted in increased use of commercial disinfectants to minimize SARS-CoV-2 transmission in indoor environments. The most widely used active ingredients within disinfectant products are quaternary ammonium compounds (QACs; e.g., benzalkonium chlorides) and alcohols [95]. Several amines (allylamine and benzylamine), and chlorides (chloropropene and benzyl chloride) were detected in the headspace air of QAC-based disinfectants (measured using a PTR-ToF-MS) [96]. These detected VOCs originated from QAC reagents, synthesis byproducts, and solvents used in disinfectants, with total emission rates of 0.16–9.6 g_{VOC} $Kg_{product}^{-1}$ [96]. Similarly, high concentrations of indoor ethanol, and monoterpenes were observed after the application of ethanol-based, and botanical disinfectants [97,98].

The temporal emission profiles of disinfectant-released VOCs were volatility-dependent, i.e., low-volatility compounds evaporated more slowly compared to high-volatility compounds [98]. Another emission feature is that the concentrations of these released VOCs were time-dependent: they first gradually increased, reaching a peak within minutes, and then decreased over time [97,98]. Consequently, the majority of VOCs inhalation intake likely occurred within the first 1-h period after disinfection. In a simulated human exposure experiment, Ding et al. estimated that 41 μg of monoterpenes and thymol were inhaled by humans when spraying thymol-based disinfectants into workplaces [99]. In summary, the use of disinfectants indoors (residence, classrooms, and offices) in unprecedented quantities during COVID-19 pandemic have increased the levels of indoor VOCs and SVOCs, which may lead to “new hazards” to humans [100,101]. This calls for an urgent need to understand the health risks of exposure to disinfectants-related VOCs.

Face masks-related VOCs. Face masks are a powerful tool for preventing the airborne transmission of SARS-CoV-2 [102]. Many synthetic chemicals are used in the manufacturing of face masks [103–105], it is reasonable to assume that some of these chemicals will evaporate to the surrounding air and become gaseous VOCs. Indeed, recent studies have detected VOCs emitted from different types of face masks (surgical masks, and N95 respirators) [106–108]. The real-time non-targeted characterization of face mask-released VOCs reveal that the most abundant VOCs are alkenes (e.g., propylene, butene, and pentene) derived from the chemicals and processes involved in mask production [108]. The initial concentration of VOCs was quite high (up to 1000 μg m^{-3}), which remained at a relatively high level within the initial 1 h (hundreds μg m^{-3}) and then decreased over time [108]. It is important to notice that wearing masks of high VOC residues significantly increased the concentration of total VOCs in the breathing zone, reflecting a relatively higher human exposure level associated with these masks [106]. Given this, it is recommended that mask manufacturing should use low VOC-emitting materials in the future [108].

4. Secondary indoor VOCs

In addition to the primary VOCs emitted from static contents, humans, human activities, and microorganism (Section 3), secondary VOCs arisen from gas-phase chemistry (Section 4.1) and heterogeneous chemistry (Sections 4.2 and 4.3) also play an important role in determining the quality of indoor air.

4.1. *Secondary VOCs formed from gas-phase reactions*

In general, gas-phase reactions have limited impact on the fate of indoor VOCs, due to the short air residence time and low oxidant levels indoors. Take isoprene as an example, at typical indoor concentrations of 4.9×10^{11} molecules cm^{-3} O_3, 1.2×10^5 molecules cm^{-3} OH radical, and 0.5–5 h^{-1} air exchange rate, reactions with O_3 and OH account for 0.4–4.5 %, and 0.9–8.6 % of the total isoprene loss indoors, respectively. Thus, only a very small fraction of isoprene will be oxidized indoors under typical indoor conditions.

Even though the lifetimes of indoor VOCs are generally not governed by gas-phase chemistry, there are expectations for this generalization. For example, some monoterpenes such as limonene and α-pinene can react 8–21 times faster than isoprene during ozonolysis [109]. Indeed, fast gas-phase oxidation chemistry was observed when cleaning an office floor with a terpene-containing cleaner [7]. It was found that during cleaning, the indoor monoterpene concentrations exceeded outdoor concentrations by a factor of 100, thus increasing the rate of O_3 reaction. Oxidation products from indoor monoterpene gas-phase reactions, including $C_{10}H_{16}O_2$ (pinonaldehyde formed from α-pinene oxidation), and $C_9H_{14}O_4$ (limonic acid formed from limonene oxidation) have been detected [7,110,111]. The gas-phase oxidation products will undergo further oxidation reactions to form highly oxygenated organic molecules which can ultimately lead to the formation of indoor secondary organic aerosol (SOA) [112].

In theory, secondary VOCs can also arise from NO_3 gas-phase reactions, which contribute to the formation of SOA in outdoor environments [113]. However, indoor NO_3 concentration is not well characterized, it is unclear whether NO_3 gas-phase reaction play a role in indoor chemistry of VOCs. In addition, gas-phase photochemical reaction can occur under certain conditions, e.g., upon direct exposure to sunlight when opening windows or upon exposure to fluorescent lights

[12,114]. Indoor oxidants such as HONO, H_2O_2, Cl_2 (released from bleach washing) may photolyze in the above-mentioned situations, forming radicals and initiating photochemical reactions, then generating secondary VOCs and indoor SOA [115–117].

4.2. Early studies of secondary indoor VOCs

In this Section, we briefly mention early studies of secondary indoor VOCs, including heterogeneous ozone-carpet, ozone-smoke, and ozone-paint reactions. Then in Section 4.3, we discuss the new advances that have occurred in the past five years. The emission characteristics of secondary VOCs are summarized in Table 2.

4.2.1. Ozone-carpet reaction

Ozone is considered to be the most important indoor oxidant [10]. Heterogeneous O_3 reactions can take place on the surface of carpets, with O_3 deposition velocity (V_d) and reaction probability (γ) of 0.016–0.064 cm s^{-1} and $\sim 10^{-5}$, respectively [118]. In 1992, Weschler et al. found that, when exposing carpets to 30–50 ppb O_3, the concentrations of 4-phenylcyclohexene, 4-vinylcyclohexene, and styrene (backing materials of carpets) decreased significantly, while the concentrations of formaldehyde, acetaldehyde, and the C5–C10 aldehydes were simultaneously formed, as measured with a GC-MS [119]. Similar phenomenon was also observed by Morrison and Nazaroff in the 2000s, which found that 2-nonenal and *n*-nonanal were the two major VOC products formed from ozone-carpet interactions, with molar formation factors of 0.16 and 0.25, respectively [120]. Interestingly, new carpets were more reactive than old ones. For example, the secondary emission rate of aldehydes for one-year-old carpet (80 μg m^{-2} h^{-1}) was higher than that of ten-year-old carpet (8–20 μg m^{-2} h^{-1}) [121]. The total aldehyde emissions from carpet surfaces are higher in summer than in winter, which is related to indoor temperature and humidity [122,123].

4.2.2. Ozone-paint reaction

Ozone can also deposit to painted materials and react with the compounds on paint surfaces to form secondary VOCs. In 1995, Reiss et al. found that carbonyls (formaldehyde, acetaldehyde, and acetone) were formed during the heterogeneous reactions of O_3 and interior latex paint, as measured with the high-performance liquid chromatography (HPLC) method [124]. The later studies reported that the V_d and γ of O_3 were 0.03–1.2 m h^{-1} and 9×10^{-8}–3.9×10^{-6}, respectively, on latex-painted walls from different aged houses and apartments [122]. In a chamber study, Gall et al. found that the V_d and total secondary carbonyl emissions were 0.32 m h^{-1} and 0–30 mg m^{-2} h^{-1}, respectively, for low-VOC painted drywall [125]. Obviously, the O_3 deposition velocity and reaction probabilities on painted walls are smaller compared to carpets. This is likely due to the lower content of unsaturated organic compounds in paints and the barrier effect of paints to O_3 transport and reactions with the underlying materials [126].

4.2.3. Ozone-smoke reaction

Tobacco smoke contains many unsaturated organic compounds, which have the potential to react with O_3. In 2001, Shaughnessy et al. examined the impact of O_3 on 18 VOCs found in tobacco smoke [127]. They found that the concentrations of aldehydes, including formaldehyde, hexanal, benzaldehyde, nonanal, and decanal, significantly increased upon exposure to $\sim$115 ppb O_3. In addition, solanesol and nicotine (the major constituents of smoke) can react with O_3 to form many products such as 4-oxopentanal, geranyl acetone, cotinine, N-methyl formamide, and nicotine-1-oxide [128–130]. The measured heterogeneous O_3-nicotine reaction rate was on the order of 10^{-5} min^{-1} (at O_3 mixing ratio 42 ppb), corresponding to a lifetime of 10 days for nicotine under typical indoor conditions [131]. Note that some of the above-mentioned reaction products are more potent irritants than their precursors in tobacco smoke, implying a potential risk to human health.

4.3. Recent advances in secondary indoor VOC measurements

4.3.1. Ozone-skin lipid reaction

The heterogeneous reactions between gas-phase O_3 and double bonds on humans and indoor surfaces have significant impacts on indoor VOCs. A long-explored indoor O_3 reaction is with human skin lipids, which are composed of many low-volatility, oily compounds including squalene, cholesterol, and fatty acid [132]. Squalene is the most abundant compound in human skin lipids, which accounts for up to 15 % of the total lipid composition and constitutes 48 % of the double bonds in skin lipid [133]. The high abundance of squalene makes it the most reactive species in skin lipids upon O_3 oxidation. It was found that when two people entered a test chamber, the O_3 concentration dropped immediately by a factor of two, while the concentrations of oxygenated VOC products from squalene ozonolysis [e.g., 6-methyl-5-hepten-2-one (6-MHO), 4-oxopentanal (4-OPA), and geranyl acetone (GA)] rapidly rose [134,135]. This oxidative chemistry has been shown to occur in genuine indoor environments as well [136,137]. During PTR-ToF-MS measurements in a university classroom, it was observed that the concentration of VOC products from skin lipid ozonolysis exhibited a positive relationship with CO_2 concentration (an indicator for human occupancy) and was anti-correlated with O_3 concentration, providing clear evidence for the occurrence of skin lipid-O_3 chemistry [137,138]. Among the O_3 oxidation products, 6-MHO and GA are the two most abundant compounds, with formation yields of 22 % and 16 % respectively, as demonstrated by the human skin-O_3 interaction experiments [139].

Intriguingly, a recent study observed oxygenated VOC products of squalene ozonolysis even after human occupants were away from the house for five days, indicating that lipid compounds can transfer to indoor surfaces and can exert chemical impacts indoors even without human presence in the house [140]. In addition to the impact on indoor VOCs, skin lipid-O_3 chemistry can also impact indoor OH concentrations. To illustrate, 6-MHO, the key product of squalene ozonolysis, can

Table 2
Summary of kinetics, gas-phase products, and condensed-phase products formed from indoor heterogeneous reactions.

Reaction type	Experimental conditions[a]	Reaction kinetics [b] (cm^3 $molecules^{-1}$ s^{-1})	Gas-phase products	Condensed-phase products	Ref.
O_3 + squalene	50 ppb O_3 + Surface-bound squalene (thickness 31 nm)	$k = 9.7 \times 10^{-16}$	Acetone, 6-MHO, geranyl acetone, secondary ozonides	Evulinic acid, succinic acid	[142]
O_3 + cholesterol	700−2000 ppb O_3 + Suspended cholesterol particle	$k = 1.0 \times 10^{-17}$	Vinyl hydroperoxides, bis-hemiacetals, ketones, and acids	/	[143]
O_3 + triolein	100 ppb O_3 +Surface-bound triolein (thickness 389 nm)	$k = 1.6 \times 10^{-16}$	Volatile C9 compounds, H_2O_2	Secondary ozonide, condensed-phase aldehyde	[148]
O_3 + oleic acid	75−675 ppb O_3 + Oleic acid	$k = 1.7 \times 10^{-16}$	9-Oxononanoic acid, volatile C9 compounds	α-Acyloxyalkyl hydroperoxides	[147, 149]
O_3 + linoleic acid	350 ppb O_3 + Surface-bound linoleic acid (thickness 200 nm)	$k = 6.2 \times 10^{-17}$	Carboxylic acids	α-Acyloxyalkyl hydroperoxide, carboxylic acids	[150]

[a] At room temperature (293−298 K).

[b] Second-order reaction rate constant (k).

further generate OH radical through gas-phase reaction with O_3 and leads to an indoor OH oxidation field around human body [141].

The ozone-skin lipid chemistry is fast, as evidenced by the measured O_3 reaction rate constants for squalene (9.7×10^{-16} cm^3 $molecules^{-1}$ s^{-1}) [142], and cholesterol (1.0×10^{-17} cm^3 $molecules^{-1}$ s^{-1}; see Table 2) [143]. The reactivity of these lipid components is likely related to their structures: Squalene contains six carbon−carbon double bonds and shows high reactivity, whereas cholesterol contains only one double bond and shows relatively lower reactivity than squalene. With the measured reaction kinetics, the estimated lifetime for surface-bound squalene is very short (35 min) when exposing to 20 ppb O_3 (a typical indoor O_3 concentration). This emphasis the importance of skin lipids-ozone chemistry when assessing indoor air quality.

The generalized mechanism for olefinic ozonolysis is shown in Fig. 2 [142,144]. When O_3 reacts with double bonds, a primary ozonide is formed which then rapidly decomposes into a carbonyl compound and a Criegee intermediate. The formed carbonyl compound is generally volatile, which can be "released" from the surface to gas-phase and becomes a gaseous VOC, as with the case of 6-MHO and GA during ozonolysis of squalene. The stability of the formed Criegee intermediate is dependent upon the RH. When the water content of the surface is low, the Criegee intermediate can undergo three different reaction channels: form carboxylic acids via isomerization mechanism (channel R1 in Fig. 2), react with carbonyl compounds to produce secondary ozonides (channel R2 in Fig. 2), and generate hydroperoxide by combining with carboxylic acids (channel R3 in Fig. 2). A recent study noticed that secondary ozonides can further react with NH_3 (also emitted from human skins) to form imines [50]. When water abundance is high, the Criegee intermediate can react with water to form α-hydroxyhydroperoxides, which can then produce aldehydes through decomposition reactions (channel R4 in Fig. 2).

4.3.2. Ozone-cooking oil reaction

Cooking oil also contains many unsaturated lipids such as triolein, oleic acid, and linoleic acid [145–147], all of which are reactive toward O_3. The O_3 reactions of these cooking oil components proceed via the mechanism similar to skin lipids (Fig. 2). When exposed to O_3, these compounds can be transformed to functionalized condensed-phase products such as secondary ozonides, aldehydes, and acids, and gaseous products such as 9-oxononanoic acid, and nonanal [148]. Given the fast triolein-O_3 reaction kinetics (1.6×10^{-16} cm^3 $molecules^{-1}$ s^{-1}) [148–150], the lifetime of a thin triolein coating indoors is short (a few hours). Zhou et al. found that the formed secondary ozonide products were thermally stable over a period of at least two days at room

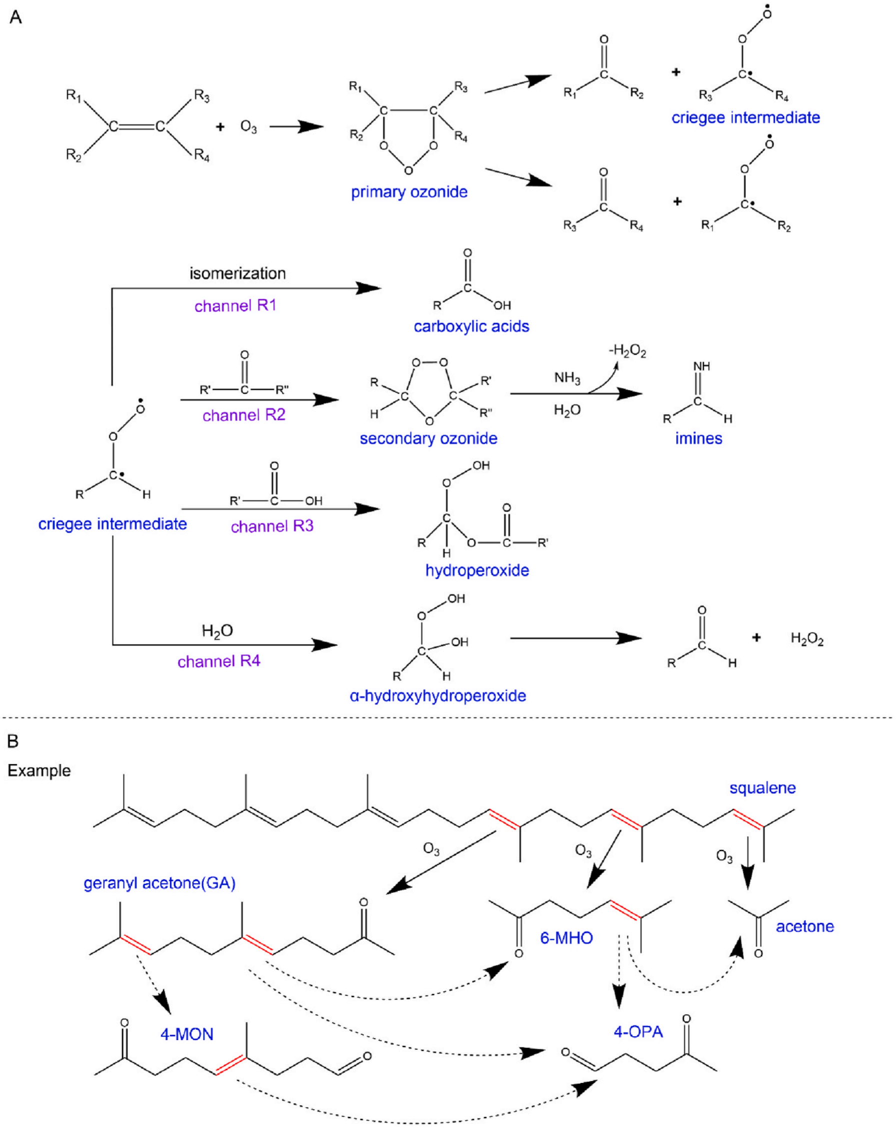

Fig. 2. (A) Reaction mechanism for ozonolysis of olefins [10,50]. **(B)** Key gas-phase products formed from squalene ozonolysis [139].

temperature, suggesting that they have the potential to accumulate on genuine indoor surfaces such as cooking areas. While secondary ozonides from ozonated oils have been applied for skin infection treatment [151], their toxicological effects are largely unknown which warrant attention. Also note that the gas-phase product nonanal has been identified as an indoor air pollutant which is linked to lung function impairment [152].

Although considerable progresses have been made recently in elucidating the O_3 transformation chemistry of human skin lipids and cooking oil constituents, there are additional questions to address in the future. First, in addition to the previously studied squalene, oleic acid, and triolein, human skin lipids and cooking oil contain many other components such as licanic acid and wax esters, for which their indoor O_3 oxidation kinetics and products distribution are unknown at present. More studies are needed to investigate the environmental fate of these compounds under indoor conditions. Second, our understanding of the influence of indoor conditions (RH, O_3 concentration, and co-existing compounds) on O_3 chemistry is incomplete. As an example, the secondary ozonides from lipids ozonolysis have the potential to react with the co-existing inorganic and organic compounds present indoors to form other products. However, studies on secondary ozonide chemistry and the associated impact on indoor air quality are rare. Third, the O_3 oxidation reactions of pure compounds (e.g., triolein) deposited on certain substrates (e.g., glasses) have been extensively studied in laboratory experiments. Note that indoor surfaces are composed of both impermeable (e.g., glass windows and stainless steel) and permeable (e.g., cotton fabric and carpet) surface materials [153], this complexity can certainly impact the heterogeneous reactivity of O_3. It would be valuable to know if the oxidation kinetics and products will be different in complex real-world environments compared to those conducted in the controlled laboratory experiments.

4.3.3. Photochemical reactions

Outdoors, condensed-phase photochemical chemistry has been extensively studied [154]. Indoors, similar heterogeneous processes can take place on certain indoor surfaces (e.g., the inner surfaces of glass windows) and contribute to the formation of secondary VOCs when exposed to direct sunlight or lights indoors. A potentially important photosensitizing agent present indoors is titanium dioxide (TiO_2), which is widely used as whitening agents in paints. A few studies have shown that upon simulated sunlight irradiation, a large suite of VOCs (e.g., formaldehyde, acrolein, and nonanal) with high emission rates, were observed to be emitted from the paint surface [155,156]. The underlying chemical mechanism is that organics in paints can react with OH radicals produced by the activation of TiO_2 and lead to the formation of secondary VOCs.

Another possible photosensitizing agent present indoors is carbonyl compounds. A recent experimental study found that when exposed to LED light, surface-adsorbed furfural (a semi-volatile carbonyl compound emitted from wood-based materials) can undergo photochemical transformation to form secondary VOCs such as maleic anhydride [157]. It remains an open question if this photochemical process plays a role in real indoor environments.

4.3.4. Air cleaners

With the increasing awareness of the importance of indoor air quality, air cleaners based on physical and chemical methods have been widely used indoors [117,158]. Physical methods are the most prevalent technology, where pollutants are physically removed through filtration or adsorption, with no by-products generated in design. However, filters have been reported to serve as a sink for ozone, and used filters removed ozone more persistently than unused ones [159].

Air cleaners using chemical methods (hydroxyl radical oxidation, and air ionization) aim to mineralize indoor VOCs by transforming these organic compounds to their oxidation products (CO_2 and H_2O), however, considerable by-products are often produced [160,161]. For example, formic acid, nitrous acid, acetic acid, iminoacetic acid, etc. formation was observed by a HR-ToF-CIMS during the use of OH generator in an office. Simultaneously, the particle number concentration reached 4000 particles cm^{-3}, with an O/C ratio of 1.3, suggesting the formation of highly oxidized SOA [162]. Air ionizers produce chemically active species (e.g., N*, O*, and OH), and a strong electric field with high-energy and short residence time to decompose VOCs [163]. Despite this, little is known about the impact of this technology on indoor air, including the formation of secondary VOCs.

Ultraviolet (UV) technology, which can induce photocatalytic oxidation to remove VOCs, is widely used indoors [162]. Although VOCs can be removed by UV irradiation, the mineralization efficiency is low (41 % on average) and is negatively correlated to VOC molecular weight. One of the unintended consequences for the use of UV-based cleaners is the formation of oxidized byproducts. For example, when UVC lights were turned on in a laboratory room, the total signals of Vocus PTR-ToF-MS and NO_3-CIMS both increased significantly, reflecting strong photo-induced secondary chemistry [161]. Specifically, the signal intensities of sulfur-containing species such as $C_6H_6O_3S$, $CHNO_3S$, and C_3H_8OS, increased by up to a factor of 10 during UVC irradiation [161]. These results indicate that unwanted indoor air quality effects may arise when using certain air cleaners, which warrants further study from the perspective of potential negative health effects.

5. Future directions

5.1. Developing advanced online and offline technique to track indoor VOCs

Although the application of online fast-response instruments such as PTR-ToF-MS greatly advances the understanding of indoor VOC behaviors, the limitations are inevitably present. For instance, one potential limitation of PTR-ToF-MS is that the isomer ions are indistinguishable. This can be improved by using hyphenated chemical characterization techniques, e.g., coupling a GC for chemical separation with a PTR or CIMS. Claflin et al. developed an in situ gas chromatograph with automatic detector switching between Vocus PTR (GC-PTR) and EI-ToF-MS (GC-EI-ToF-MS) for isomer-resolved molecular identification of indoor VOCs, including hydrocarbons, oxygenates, and nitrogen-containing compounds [164]. Similarly, Bi et al. coupled a GC (TAG) simultaneously to a flame ionization detector and CIMS for isomer-resolved measurements of organic compounds present in both gas- and particle-phase [165,166]. These hyphenated techniques provide isomer resolved information of indoor air composition, while leading to a reduced time resolution [167]. Therefore, researchers should make a balance of their choice.

Furthermore, techniques based on higher resolution mass spectrometry have been used in the past few years in indoor air studies. GC-Orbitrap-MS was used for non-targeted screening of VOCs in a museum in China [168]. Although currently, the samples are analyzed offline after collection, in theory, this method can be extended to online measurements if combined with techniques such as SV-TAG. This will enable time resolved measurements with higher mass resolution. Also, other techniques such as two dimensional GC can enhance the separation ability for complex samples [169].

5.2. Exploring the unknowns (unknown VOCs and unknown sources)

With the rapid growth of the global chemical industry in recent years, numerous synthetic chemicals are produced and used indoors. Many of these synthetic chemicals will inevitably be released to indoor air (through volatilization and abrasion) and become gaseous VOCs. Despite the progresses made in indoor VOCs identification over the past decade, a knowledge gap exists on the emissions of indoor emerging pollutants. For example, a recent study detected dibasic esters (DBEs), a previously unrecognized indoor VOCs, in the indoor air of new apartments in Beijing, China [94]. The source of these newly identified DBE compounds is attributed to the coatings of furniture. Also, although LCDs have been used in daily life for decades, only recently it was demonstrated that they can emit some "unusual" chemicals such as liquid crystal monomers, to indoor air [52]. These results underscore the need to evaluate VOCs release from many more materials and devices used indoors than what have been done to date, which is particularly important given the increasing use of synthetic chemicals in new buildings. Ultimately, this will contribute to a more complete understanding of the chemical composition of the indoor air we breathe.

Another aspect of the "unknowns" for indoor VOCs is their source identification. For example, a previous study indicated that the concentration of indoor isoprene in an unoccupied house in the USA (∼1 ppb) was much higher than that in outdoor air (0.2−0.5 ppb) [11]. The known sources of indoor isoprene include human breath, and liquid crystal displays. In vacant houses, the isoprene emitted from LCDs alone cannot explain the observed isoprene level indoors given the low emission rate of LCD-released isoprene [52]. Thus, there must be other missing sources of indoor isoprene. This calls for further studies on the source appointment of indoor VOCs including isoprene.

5.3. *Toward a better understanding of oxidation chemistry on indoor surfaces*

A large fraction of indoor VOCs is now known to be formed from indoor surface chemistry. On the other hand, VOCs themselves are found to be "semi-volatile" indoors, exhibiting similar behavior as SVOCs, which are "stored" in indoor surface reservoirs [11,170]. These surface-bound VOCs and SVOCs undergo continuous modification with exposure to indoor gaseous oxidants such as O_3, OH, NO_3, HONO, etc. [171]. Indoor environmental conditions including temperature, humidity, air exchange rate, surface properties (e.g., pH, chemical composition, and structure) can affect the heterogeneous reactions of surface-sorbed VOCs and SVOCs, ultimately leading to the generation of secondary products. To mitigate the risk of exposure to indoor VOCs, a better understanding of surface oxidation chemistry is warranted, which will provide key information for source identification of indoor VOCs and can inform future development of indoor air quality control measures. In particular, existing studies on surface oxidation chemistry are mostly conducted in laboratory (i.e., in the simulated environments), research on realistic indoor surfaces and in real indoor environments needs to be explored.

5.4. *Elucidating the health impact of indoor VOCs*

With the discovery of emerging air pollutants indoors, a key question emerges: will these newly identified compounds pose new health risks to humans? To answer this question, fundamental knowledge of the emission rates, the gas-surface partitioning behaviors, the indoor transformation chemistry, the environmental factors affecting the human chemical exposure, and the toxicity of these emerging pollutants is required. However, such information is currently lacking. For example, little is known about the toxicological effects of the newly discovered liquid crystal monomers [172]. Also, the indoor presence (e.g., in air, suspended particles, and dust) of QACs with varying chain lengths remains largely unknown [101]. Such a lack of information hampers an accurate health risk assessment for these chemicals of emerging concern.

In addition, it should be noted that for many well-studied indoor pollutants, their toxicity is well known, but the toxicity of their indoor transformation products may be not. This is because the reaction products of many synthetic chemicals used indoors are largely unknown, making it very challenging to evaluate the resultant health effects. For example, a recent study found the heterogeneous reactions of bisphenols (a well-studied indoor pollutant) with indoor HONO can lead to the formation of nitrated bisphenols, which are previously unidentified toxic pollutants with widespread presence indoors [173]. Also, the information on the toxicity of most squalene-O_3 reaction products and the corresponding exposure risk is still lacking. This highlights the importance of considering the reaction products formed through secondary chemistry (including both the condensed-phase products and gas-phase VOC products) when assessing the health risks associated with indoor pollutants.

Finally, indoors, humans are exposed to a "cocktail" of numerous VOCs. Previous toxicity studies mainly focus on the health effects of individual VOC, limited information is available regarding the health effects of the combined VOC mixture. Further study is warranted to investigate the mixture effects in complex real-world indoor environments. In particular, given the long-term human exposure to indoor VOCs, in vitro and in vivo chronic toxicity studies of these VOCs are urgently needed.

CRediT authorship contribution statement

Wei Liu: Data curation, Writing – original draft, Formal analysis, Investigation, Methodology. **Li Zhou:** Data curation, Formal analysis, Investigation, Methodology, Writing – original draft. **Wenting Yuan:** Data curation, Formal analysis, Investigation, Methodology, Writing – original draft. **Ling Ruan:** Investigation, Methodology. **Xinkai Wang:** Investigation, Methodology. **Yucong Guo:** Investigation, Methodology. **Zhouqing Xie:** Investigation, Methodology. **Qifan Liu:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Supervision, Writing – original draft, Writing – review & editing, Resources. **Chen Wang:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Resources, Supervision, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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