Review

Recent advances on SOA formation in indoor air, fate and strategies for SOA characterization in indoor air - A review

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HIGHLIGHTS

- Secondary particle formation in an indoor environment was described.
- Analytical techniques applied to study secondary particle formation were presented.
- The particle phase indoor is composed of particles emitted from various sources.
- Current pandemic situation for sure increased the importance of indoor air quality.
- Most relevant literature data on secondary organic aerosol formation were presented.

GRAPHICAL ABSTRACT
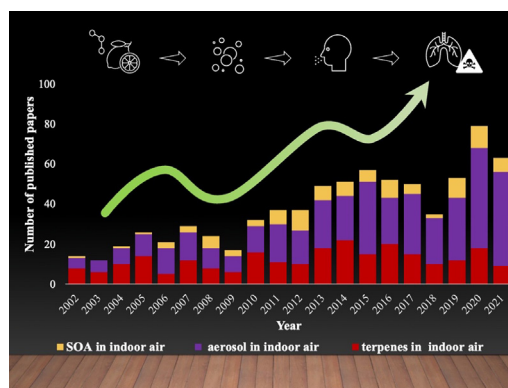
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ABSTRACT

Recent studies proves that indoor air chemistry differs in many aspects from atmospheric one. People send up to 90 % of their life indoors being exposed to pollutants present in gas, particle and solid phase. Particle phase indoor is composed of particles emitted from various sources, among which there is an indoor source – secondary chemical reactions leading to formation of secondary organic aerosol (SOA). Lately, researchers' attentions turned towards the ultrafine particles, for there are still a lot of gaps in knowledge concerning this field of study, while there is evidence of negative influence of ultrafine particles on human health. Presented review sums up current knowledge about secondary particle formation in indoor environment and development of analytical techniques applied to study those processes. The biggest concern today is studying ROS, for their lifetime in indoor air is very short due to reactions at the very beginning of terpene oxidation process. Another interesting aspect that is recently discovered is monoterpene autooxidation process that leads to HOMs formation that in turn can influence SOA formation yield. A complex studies covering gas phase and particle phase characterization, but also toxicological studies are crucial to fully understand indoor air chemistry leading to ultrafine particle formation.

Contents

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1. Introduction

Air pollution is currently recognized as one of the factors, which contribute to several adverse health impacts such as chronic obstructive pulmonary disease, lower respiratory illness, ischaemic heart disease, lung cancer (Burnett et al., 2014), birth defects (Padula et al., 2013) or neurodegenerative disorders (Moulton and Yang, 2012). Moreover, it has been proved by a global atmospheric chemistry model that there is an evident link between outdoor air pollution, particularly urban organic aerosols, and premature human mortality (Lelieveld et al., 2015). Indoor air chemistry may be considered as a subdiscipline of atmospheric chemistry field. It is a known fact that these two environments substantially differ from each other (by environmental conditions such as temperature and availability of sunlight, surface-to-volume ratio, concentrations of organic compounds) and so does the specifics of their pollution (Weschler and Carslaw, 2018). For these reasons, indoor environments should be investigated separately as individual systems. Gaining an accurate knowledge on factors which govern human exposure in indoor environments is fundamental, taking into consideration that in developed countries people spend indoors up to 90 % of their time (Long et al., 2001), or even more if the circumstances are exceptional (e.g. ongoing pandemic). Worth emphasising is also the fact that according to the World Health Organization (WHO), indoor air pollution contributes annually to the deaths of 3.8 million people (WHO, 2021).

Spending most of their time indoors, people are nearly constantly exposed to a variety of pollutants. These are (i) present in gaseous and condensed phase volatile organic compounds (VOC), (ii) present in condensed phase semi-volatile organic compounds (SVOC) and particulate matter (PM) and (iii) pollutants present in solid phase (house dust). Numerous epidemiological and related studies revealed the direct link between deterioration of human well-being and adverse health effects with fine particles present in breathing air (Rohr, 2013; Baltensperger et al., 2008; Lin et al., 2016; Chowdhury et al., 2018).

Typically, the condensed phase of indoor air is composed of particles that penetrate from outdoors, particles emitted indoors, and particles generated indoors via secondary reactions (called secondary organic aerosol; SOA) of reactive organic compounds deriving both from indoors and outdoors.

Lately, the attention has been shifted towards respirable fractions of particles, with particular regard to ultrafine particles (aerodynamic diameters smaller than 0.1 μm), where there are still numerous gaps in knowledge regarding epidemiological and toxicological understanding. These particles are considered to pose the most serious threat to human health, since they can reach gas exchange regions of lungs or even penetrate these regions to reach the circulatory system. Moreover, they have a greater surface area to mass ratio in comparison to larger particles, providing a larger area to adsorb and further transmit potentially toxic chemicals or pathogens and interface with pulmonary surfaces (Heinzerling et al., 2016). Secondary organic aerosol has been evidenced as an important source of ultra-fine particles in indoor environments.

Taking into consideration the time people spend indoors it is very likely that total human exposure to fine particles strongly depends on their presence and transformations in indoor environments. Increased awareness of indoor air quality IAQ importance led to the number of studies aimed at characterization of SOA generation indoors and its contribution to particle concentration within residences. There has been also a significant development in instrumental solutions dedicated to tracking the changes in gaseous phase leading to SOA formation as well as studying the dynamism of this process. However, there is still much to be done to fully characterize the processes that occur indoors in gaseous and particle phase and via gas-surface partitioning. Environmental scientists strongly emphasize the need to understand the multi-phase chemistry shaping IAQ and the dependence between outdoor pollution and levels of defined air contaminants indoors (Abbatt and Wang, 2020).

Published so far papers concerning indoor air chemistry and quality have been summarized in a number of review papers (see Table 1). It can be seen that in the past five years the major emphasis in IAQ-related literature has been put on characterization of real indoor environments, every day sources of indoor air pollutants and strategies towards indoor air purification and IAQ improvement. However, there is an evident lack of a review paper that summarizes specifically the most recent studies on SOA presence in indoor air. This paper is a comprehensive summarizing of the most relevant literature data on SOA formation, fate and strategies for its characterization in indoor air published in the past decade. The paper refers both to chamber and field studies as well as to some contradictory data, to give the reader a broad perspective in the field.

A comprehensive literature search was performed to identify relevant articles indexed in Scopus, SciFinder, Web of Science together with a meta-search in Google Scholar based primarily on the following key words: 'indoor air', 'SOA', 'HOM', 'ROS', 'terpenes', 'particles', 'secondary organic aerosol', 'reactive organic compounds'. References contained in the identified articles were also assessed. This review is based on the data gathered in this query and also on the authors' personal experience in the field.

2. SOA formation and fate indoors

The first observation of SOA formation in indoor environments has been reported by Weschler and Shields in 1999 (Weschler and Shields, 1999). Authors conducted experiments in identical office rooms depicting typical indoor setting. For the experiment, a known concentrations of selected terpenes (D-limonene, α-terpinene, or a terpene-based cleaner mainly containing α-pinene) and ozone were introduced into one of the offices, while the other one served as a control. Authors determined particle formation and redistribution by using eight-channel optical particle counter. The results allowed authors to conclude that ozone and terpene reactions can be a significant source of sub-micron particles in indoor air.

Since then, the main emphasis in this field of research has been put on reactions of oxidation (particularly ozonolysis) of reactive organic compounds (such as terpenes, terpenoids, and their oxidation products) in indoor environment that occur in a comparable time or faster than the air exchange rate. Gas-phase oxidation is followed by generation of multi-functional compounds and particle-phase transformations. As a result, SOA is formed.

Table 1

Review papers published in 2015–2020, summarizing knowledge about indoor air chemistry and quality.

Title	Scope of the review (taken from the Abstract/Introduction section)	Year of publication	Reference
A review of indoor air treatment technologies	• Treatment techniques to improve IAQ: mechanical and electrical filtration, adsorption, ozonation, photolysis, photocatalytic oxidation, biological processes, membrane separation. • Combined purification technologies and innovative alternatives that are currently under research and have not yet been commercialized.	2015	(Luengas et al., 2015)
Indoor air quality and its effects on humans—A review of challenges and developments in the last 30 years	• Trends in the society that renders IAQ a priority to be addressed. • Evaluation of their implications on IAQ, the effectiveness of solutions in mitigating them and the challenges that need to be addressed.	2016	(Tham, 2016)
Towards practical indoor air phytoremediation: A review	• Active air phytoremediation technology. • Available botanical biofiltration systems with indicating the areas in need of development.	2018	(Pettit et al., 2018)
A review of semi-volatile organic compounds (SVOCs) in the indoor environment: occurrence in consumer products, indoor air and dust	• Main classes of SVOCs used in consumer products and present in indoor air and dust. • The dependence between information already available for chemicals in indoor air and dust and the source of such chemicals in products.	2018	(Lucattini et al., 2018)
A state–of–the-art review on indoor air pollution and strategies for indoor air pollution control	• The state-of-the-art of the indoor air pollution problem. • Prevention strategies, the recent advances in physical-chemical and biological technologies for indoor pollutants abatement.	2021	(González-Martín et al., 2021)
A review of standards and guidelines set by international bodies for the parameters of indoor air quality	• Summarizing of the main standards and guidelines related to key indoor air pollutants and levels of thermal comfort developed by different agencies around the world.	2015	(Abdul-Wahab et al., 2015)
A comprehensive review on indoor air quality monitoring systems for enhanced public health	• The use of wireless technologies for the development of cyber-physical systems for real-time monitoring. • Microcontrollers used for system designing and challenges in the development of real-time monitoring systems. • New ideas and scopes in the field of IAQ monitoring.	2020	(Saini et al., 2020)
A systematic review of possible airborne transmission of the COVID-19 virus (SARS-CoV-2) in the indoor air environment	• Summarizing of experimental studies related to airborne transmission of SARS-CoV-2 in indoor air environment. • Recommendations for IAQ improvement in the context of SARS-CoV-2 spread in indoor environments.	2020	(Noorimotlagh et al., 2020)
Human exposure to indoor air pollutants in sleep microenvironments: A literature review	• Exposure characteristics, the biological and chemical composition of mattress dust, chemical emissions from mattresses and bedding materials. • The use of personalized ventilation and air cleaning systems for beds.	2017	(Boor et al., 2017)
Indoor Chemistry	• The importance, ubiquity, and complexity of indoor chemistry. • The sources of indoor air pollutants and their chemical reactions in the air and on surfaces. • The impacts of human occupants, who act as sources and sinks of indoor chemicals.	2018	(Weschler and Carslaw, 2018)
The atmospheric chemistry of indoor environments	• Recent advances in the understanding of the different partitioning and reactive processes that occur indoors in contrast to what occurs outdoors.	2020	(Heinzerling et al., 2016)
Indoor air quality of everyday use spaces dedicated to specific purposes—a review	• Air quality in everyday use spaces dedicated to specific purposes which are integral parts of residential buildings, such as kitchens, basements, and individual garages.	2018	(Marć et al., 2018)
Indoor hospital air and the impact of ventilation on bioaerosols: a systematic review	• The concentrations and compositions of indoor bioaerosols in different areas within hospitals and the effects of different ventilation systems.	2019	(Stockwell et al., 2019)
Does the ubiquitous use of essential oil-based products promote indoor air quality? A critical literature review	• The impact of essential oil-based household products on indoor air quality – household products composition, experimental methodologies for terpenes monitoring, dynamics of terpenes emission, fate of terpenes indoors.	2020	(Milhem et al., 2020)
Indoor air chemistry: Terpene reaction products and airway effects	• Gas-phase ozone-initiated mono- and sesqui-terpene chemistry effects on residents in office-like environments. • A review on how terpene-related surface reactions may alter the perceived air quality.	2020	(Wolkoff, 2020)
Ultrafine particles: A review about their health effects, presence, generation, and measurement in indoor environments	• A summary of health effects, generation, and measurement of ultrafine particles in indoor environments. • Discussion on issues concerning ultrafine particles measurement and assessment.	2022	(Marval and Tronville, 2022)

Reactive organic compounds in indoor air undergo reactions with oxidants like ozone, hydroxyl radicals and nitrogen oxides. The mechanism of SOA formation via gas-phase reactive organic compound oxidation is initiated by the generation of organic radical, which is followed by multiple addition of O_2 and isomerization. This results in the formation of hydroperoxy functional groups. As a consequence of these reactions, a low-volatility species are formed, which further condense to produce SOA (Ehn et al., 2014). The condensed-phase transformations are initially dominated by nucleation and condensation processes. Within nucleation, semi-volatile compounds present in gas phase form clusters of few nm in size. There can be homo- and heteromolecular nucleation, depending on the types (correspondingly the same and different) of interacting semi-volatile species. Nucleation depends predominantly on vapour pressure and affinity of interacting molecules. Residence time of particles formed via nucleation is short (few minutes). Depending on the size and the surface tension, compounds forming nanoparticles may evaporate to gas phase or

these nanoparticles may further undergo condensation and coagulation, which leads to particle growth. Condensation of semi-volatile vapors on pre-existing particles allows them to grow in size. This process depends on the size of pre-existing particles, diffusion coefficient and vapour pressure of species present in gas phase. Condensation affects particle size, while particle number concentration remains constant. Coagulation is a process of collision and aggregation of particles, which leads to the increase of particles size and decrease in particle number concentration. It depends on particles concentration and their excitement (thermal, dynamic). Described processes are presented in Fig. 1 (Hallquist et al., 2009; Kroll and Seinfeld, 2008; Kavouras and Stephanou, 2002; Westervelt et al., 2013).

A vast number of studies focused on ozone reactions with various terpenes and terpenoids were conducted and published in recent years, therefore the information on terpene ozonolysis reaction mechanism are well documented. The scheme of transformations of selected terpenes
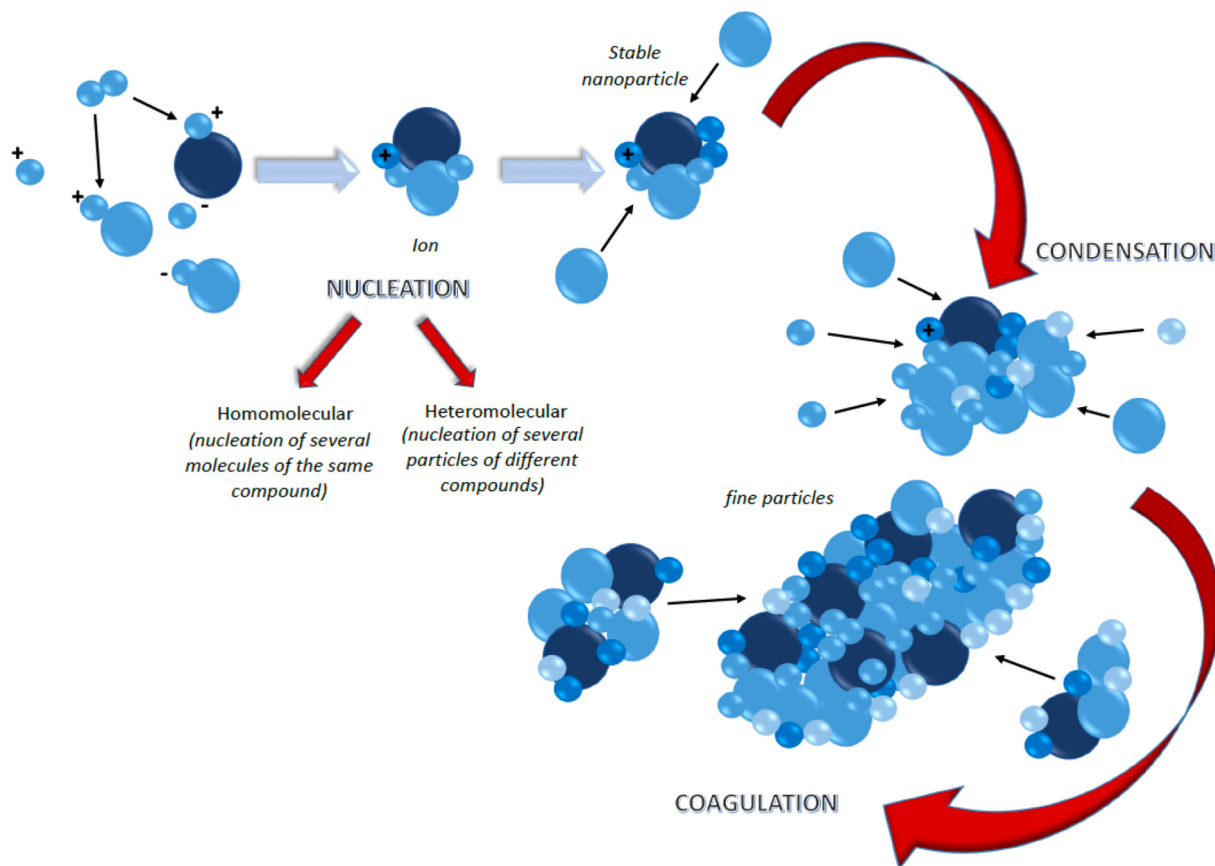
Fig. 1. Major processes leading to SOA formation: nucleation, condensation and coagulation.

(monoterpene limonene, and triterpene squalene) initiated by ozone as oxidant is presented in Figs. 2 & 3.

Ozone is believed to be the most abundant oxidant in indoor air (Xiong et al., 2019). It's presence indoors may be a result either of the transport from outdoors in a concentration ranging from c.a. 20 to 70 % of the atmospheric level (Nazaroff and Cass, 1986; Charles J. Weschler, 2000) or of in-place emission from photocopiers or air purification devices (Allen et al., 1978). Based on some literature data, ozone concentration

indoors can be found in a range from several to several dozens of vppb. As an example, Weschler et al. reported ozone concentration in the office rooms to be ~17–37 vppb (among 6 measurement days in California, USA) (Weschler et al., 1994). In the review article published in 2018 by Salonen et al., one can find the information about ozone concentration in office building across USA, Europe and Asia, reported over 1973–2017. Reported values cover range ~ 2–41 vppb (Salonen et al., 2018). Recently, in 2022, Nazaroff et al. have published a review paper containing a data on
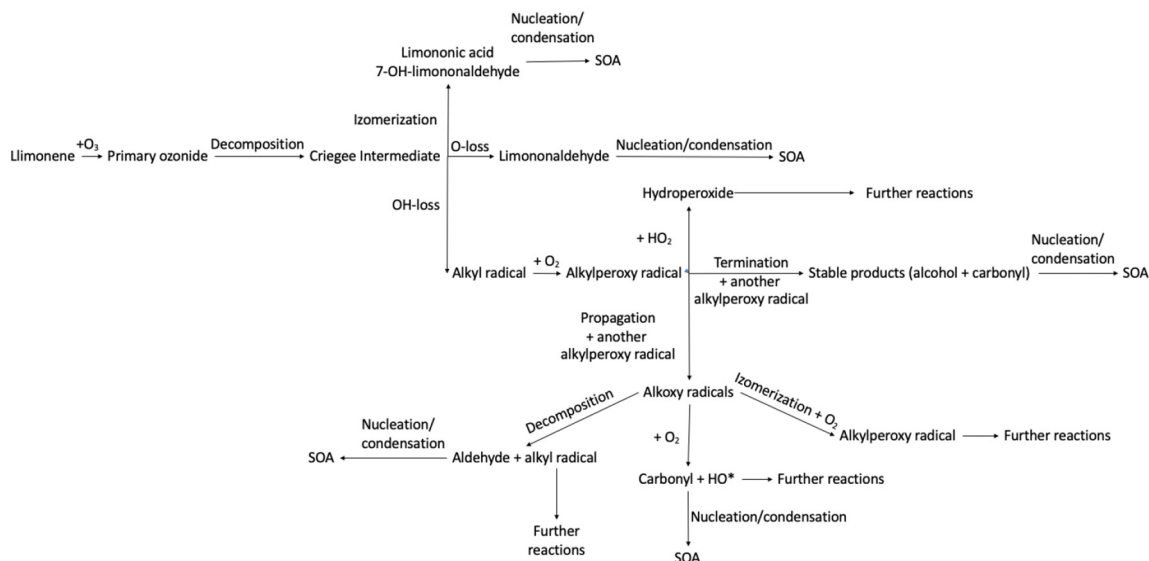

Fig. 2. Chemical transformations initiated by limonene ozonolysis leading to SOA formation, based on M.L. Walser et al. (Walser et al., 2008).
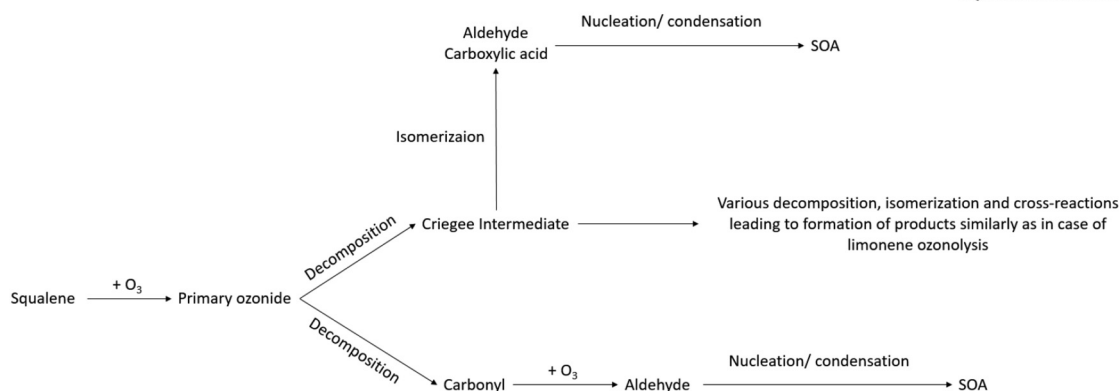
SQUALENE OZONOLYSIS MECHANISM

Fig. 3. Chemical transformations initiated by squalene ozonolysis leading to SOA formation in study where squalene directly from human skin was scrubbed onto glass wool and exposed to 100 ppb ozone, based on D.R. Fooshee et al. (Fooshee et al., 2015).

ozone concentrations in residencies located in USA, Canada, Europe and Asia over the period 1981–2017. Indoor ozone concentration range was ~1–21 vppb (Nazaroff and Weschler, 2022)."

The presence of terpenoids indoors depend mostly on emissions from air fresheners, perfumes, cleaning agents and wood products (Waring et al., 2011) or during common human activities such as peeling an orange. Interestingly, orange peeling in an enclosed space showed SOA yield around 50 times higher than using a limonene floor cleaning agent (Arrhenius et al., 2008). The average concentration of D-limonene, which is known as predominant indoor terpenoid, may reach up to 50 ppb indoors (Weisel et al., 2005), however such incidental concentrations as 1000 ppb during above mentioned products use have been also reported (Singer et al., 2006). Ozone/terpenoids reactions are believed to be initiated by electrophilic addition of ozone on the terpenoid double bond, which leads to the formation of excited primary ozonide. This product rapidly undergoes decomposition into excited carbonyl oxide called Criegee intermediate, which is highly reactive. It can be stabilized by the collisions with the other gaseous particle (Atkinson, 1997; Criegee, 1975). Both excited and stabilized forms of Criegee intermediate may react to produce OH radicals, alkylperoxy radicals (RO_2) or undergo further complex chemical transformations, which result in the formation of such chemical species as ketones, diketones, keto-aldehydes, hydroxy-ketoaldehydes, keto-carboxylic acids, dicarboxylic acids, formaldehyde (Kim et al., 2010; Koch et al., 2000) and also acrolein, formic acid and acetic acid (Arrhenius et al., 2008; Wolkoff et al., 2008). The species characterized by low volatility contribute to the formation of SOA. This fate is most likely for multifunctional compounds with high molecular mass of 150–300 g mol^{-1} and with ambient vapour pressures below 0.1 Pa (Barley and McFiggans, 2010). Next to already mentioned D-limonene, other well-studied and documented terpenoids proved to be the precursors of SOA indoors are α- and β-pinene, α-terpineol, linalool and dihydromyrcenol (Pullinen et al., 2020; Palmisani et al., 2020). Although several studies revealed that ozone in the reaction with D-limonene produces more SOA particles than with α-pinene (Sarwar and Corsi, 2007), which is a consequence of two (*endo*- and exocyclic) double bonds in the structure, it is α-pinene that exhibits higher aerosol formation potential in terms of monoterpenes reactivity (Leungsakul et al., 2005a). Apart from monoterpenes, gas-phase reactions leading to SOA formation may derive from such precursors as cigarette smoke (Wang et al., 2018) and cooking oils (Liu et al., 2018). Ozonolysis of surface-sorbed unsaturated compounds (e.g. squalene on skin (Wang and Waring, 2014) or surface-sorbed D-limonene (Waring and Siegel, 2013)) has been also proved to yield SOA. It has been reported that surface ozone/D-limonene reactions promoted nucleation more than gas-phase reactions and that this way of SOA formation is mostly significant in case of surfaces of low original reactivity with ozone, i.e. glass, sealed materials or smooth metals (Sarwar and Corsi, 2007). Another possible source of SOA particles indoors is desorption of SVOC from surfaces within high-temperature related household activities such as

using cooking utensils, stovetops or radiators. Accumulated SVOC may desorb upon heating from the mentioned surfaces. This process forms the plume of air which rises and cools and leads to the supersaturation and nucleation to generate SOA (Weschler and Carslaw, 2018).

SOA formation rate is strongly related to indoor environmental conditions, mainly relative humidity (RH). Moreover, the RH increase enhances the sorption of polar compounds to indoor surfaces, what makes the reactions on surfaces more probable. Also, the course of transformations of highly reactive intermediates of SOA formation may be substantially different in dry/moist conditions. It has been reported, that ozonolysis of monoterpenes is influenced by RH - higher RH correlates with increased particle formation due to nucleation (Rösch et al., 2017).

Although the oxidation of reactive organic compounds (particularly monoterpenes) by ozone is best documented process that initiates SOA formation indoors, the role and consequences of reactions with hydroxyl (OH) and nitrate (NO_3) radicals should not be neglected and therefore further investigated. Apart from air exchange, OH and NO_3 formation may occur primarily from O_3 reactions with alkenes and nitrogen dioxide (NO_2), respectively. Outdoor-to-indoor transport of OH radicals is not considered as a strong indoor source because of their short lifetimes (Carslaw, 2007). Hydroxyl and nitrate radicals may react with many different VOCs, unlike O_3, which reacts only with alkenes (Waring and Raymond Wells, 2015). Hydroxyl radicals are believed to affect indoor terpenoid chemistry to a lesser extent than O_3, however there are studies in which equal contribution of these two oxidants to D-limonene conversion has been suggested. Nitrate radical should be of particular concern, since it can relatively fast oxidize the reactive species and it is not vulnerable to photolysis indoors (Nazaroff and Goldstein, 2015). It has been noted that concentration ratio of [O_3]/[NO] around 1 was found to produce the highest mass of SOA and reactive oxygen species (ROS). Therefore, SOA and reactive oxygen species could be formed when NO concentration is low enough in a way that part of NO_2 could be converted into nitrate radicals, however it would not be so efficient as in case of ozone-only oxidation. When indoor NO concentration is significant, infiltrated O_3 will most probably be consumed in the process of NO_2 formation. The lowest concentration ratio of [O_3]/[NO] was found to produce the highest SOA density. Additionally, SOA mass yields positively depend on [O_3]/[VOC] ratio without NO_2. Introduction of NO_2 causes increase in SOA yield when [O_3]/[VOC] > 2. While [O_3]/[VOC] < 2, a negative effect for SOA formation was observed (Liu and Hopke, 2014). Next to alkyl, alkoxy and peroxy radicals, NO_3/VOC reactions result in oxygenated compounds with organic nitrate groups in their structure (Ham, 2013). Nitrated species have been hypothesized next to peroxide ones to dominate SOA since they are likely to condense onto existing particles.

SOA particles formation occurs via homogenous nucleation as well as condensation on pre-existing particles, which is governed by partition mechanisms (Destaillats et al., 2006). Nucleation is hypothesized to occur

just after the beginning of the reaction initiating the particle formation. It affects particle number concentration. Partitioning in turn impacts particle mass formation and may occur any time the reaction progresses (Waring et al., 2011).

Recently it has been discovered that SOA generation may be enhanced by rapid formation of species characterized by extremely low volatility, called highly oxygenated organic molecules (HOMs) or extremely low volatility organic compounds (ELVOCs). Oxidation of VOCs generates peroxy radicals (RO_2), which lifetime is sufficiently long to reach high concentrations and undergo multiple auto-oxidation reactions, resulting in multifunctional HOMs (Peräkylä et al., 2020; Bianchi et al., 2019). These species are hypothesized to highly influence new particle formation through nucleation and growth and if sorbed, they will not re-evaporate from a particle or surface (Ehn et al., 2012). According to model studies, SOA composition may be dominated by HOM species in some certain indoor conditions, such as high levels of nitrate radicals (Kruza et al., 2020).

SOA fate is highly related to air exchange rate, which means that its further transformations are mainly limited by indoor-to-outdoor transport. This in turn strongly relies on the type and efficiency of ventilation systems. A certain role is also played by air purifiers equipped with filters. SOA particles may be also deposited on indoor surfaces with a formation of organic films (Weschler and Nazaroff, 2017). It has been also reported that potted plants, especially those with broad leaves, variegated surface and dense (e.g. *Chamaedorea elegans, Peperomia jayde*), may help in reducing ultrafine particles concentration indoors (Stapleton and Ruiz-Rudolph, 2018).

3. Major threats related to ultrafine and fine particles presence in breathing air

Human beings predominantly breathe an indoor air, which is a consequence of spending most of their time in enclosed spaces (e.g., homes, schools/working environments, shopping malls, hospitals, gyms etc.). For this reason, any indoor air pollutant should be considered as a threat to human health.

SOA fraction is mainly composed by ultrafine particles of diameters smaller than 100 nm and has been also recognized as a source of fine PM2.5 particles. It is still under investigation what is the contribution of SOA to indoor PM2.5 concentration. There were model-based estimations which revealed that this contribution is little, at a level of <3 % (Ji and Zhao, 2015). However, this definitely needs further investigation in real indoor environments.

The potential for ultrafine particles to cause adverse health effects is great, but their precise role in causing specific illnesses needs further investigation. So far, inhalation of these particles have been linked to worsening asthma, cardiovascular diseases, hypertension, diabetes, cancer lesions, central nervous system dysfunction as well as low birthweight (as a consequence of pregnant woman exposure) (Schraufnagel, 2020).

Because of their ultra-small size, ultrafine particles remain suspended in the air longer and may easily penetrate human body barriers to alveoli and translocate across alveolar epithelial cells by diffusion into the bloodstream and further to all organs. Remained longer in the lungs, ultrafine particles cause pulmonary inflammation. The finer the inhaled particles, the stronger and more persistent inflammation occurs. This is another mechanism by which these particles harm organs other than lungs – as a result of inflammation, inflammatory mediators are spread to distal organs (Schraufnagel, 2020).

Toxicity of ultrafine particles is likely related to their large surface-to-mass ratio. This property provides the ability to transfer different species of chemicals adsorbed in large amounts per unit mass. SOA may pose a threat to human health by acting as sorptive reservoirs for ROS formed during oxidative processes or semi-volatile organic compounds (SVOC) emitted indoors, such as plasticizers, pesticides or polycyclic aromatic hydrocarbons (PAHs) (Waring, 2014). Also their charge, surface reactivity, solubility and hydrophobic/hydrophilic properties determine particles interaction with biological tissues (Moreno-Ríos et al., 2022).

Particles suspended in the indoor air that consist of bacteria, viruses, fungi and/or their metabolites as well as plant pollen and fragments of plant tissues are categorized as indoor bioaerosol. Recently this fraction of indoor aerosol got particular significance because of possibility to transmit coronavirus SARS-COV-2 in enclosed spaces at distances >2 m. Since the aim of this review is to summarize the most recent findings on IAQ in the context of indoor aerosol particles, SARS-COV-2 issue and relation between its transmission and airborne particles could not be omitted here. It is suspected that particulates create a suitable environment for virus transmission and, by inducing lung cell inflammation, increase the susceptibility to COVID-19 and severity of illness course (Domingo et al., 2020). It has been reported that coronavirus SARS-COV-2 has been detected in aerosol droplets of 2.5 μm diameter or smaller, which are likely to remain suspended in indoor air for even >2 h (Allen and Marr, 2020). It may be then assumed that this virus (similarly to other already studied viruses) can survive well in suspended aerosols. Obviously, the major source of these "virus-rich" aerosols is respiratory secretion from an infected person. However, the presence of viral RNA has been reported on the surfaces of an air exhaust outlet and fan, which could not derive from an infected person.

The potential for ultrafine particles to cause adverse health effects is great, but their precise role in triggering many illnesses is still under investigation. Because they are produced intensively and redistribute rapidly, incidental exposure to them is very common. Therefore, extended research on a global scale in this field is hindered. To fill this gap in the knowledge, standardized measurement protocols are definitely needed.

4. Indoor SOA investigation – strategies and instrumental solutions

Investigation of indoor air chemistry can be done by conducting two types of studies – field and chamber. Field studies are most commonly aimed at determination of concentration of analytes, or at investigation on how certain activities/emission sources influence analytes concentration in real environment (Pagonis et al., 2019; He et al., 2016). In case of terpene indoor chemistry, literature data proof that field studies are most commonly focused on investigation of gas phase, more precisely determination of terpene and oxidation product concentration (Schlink et al., 2016; Mandin et al., 2017). Particle phase is not so commonly investigated during indoor field studies (Cincinelli et al., 2016; Rösch et al., 2015). It is also relatively rare that both gas and particle phase are investigated at the same time. Chamber studies are most aimed at investigation of terpenes oxidation chemistry in environmental conditions that mimic real indoor environment. Application of reaction chamber (that generally have from few to few dozen m³ (Youssefi and Waring, 2014; Yu et al., 2011; Nørgaard et al., 2014)), facilitates control of environmental conditions, which would be much more difficult in real indoor environment. Chamber experiments allow to easily control parameters that influence terpene/terpenes oxidation reactions: temperature, humidity, substrate concentration or reaction time. Thanks to the ease of control, one may add to the experiment some additional factors influencing terpene oxidation reactions, like nitrogen oxides, OH scavengers or seed particles (Chen et al., 2017; Gallimore et al., 2017b; Hu et al., 2017).

4.1. Chemical analysis of SOA precursors present in indoor gas phase: measurement techniques and examples of their implementation

There is a range of analytical techniques for investigation of indoor gas and particle phase that developed much over past 20 years. At the beginning of XXI century most of the studies applied off-line techniques for measurement of terpene substrates. Concentration of gaseous analytes was determined with application of passive or active sampling on a sorbent (mostly Tenax TA®), which was followed by thermal desorption and qualitative and quantitative analysis with gas chromatography (TD-GC-FID or TD-GC–MS) (Klenø et al., 2006; Toftum et al., 2008; Arrhenius et al., 2008). However, application of those techniques has few limitations, such as: receiving information only about temporary mean concentration (related to the sampling time) of gaseous analytes, sometimes problematic

choice of sorbent due to different polarity of analytes, difficulties with determination of oxidation products concentration due to their short residence time (and therefore, very low concentration). Most commonly, this type of studies was focused on investigation of the potential of SOA formation by manipulating the environmental parameters such as NO_x, O_3 and terpene concentration ratios in chamber studies. Usually, this kind of studies were supported by modeling gas-phase reactions (Arrhenius et al., 2008; Leungsakul et al., 2005b).

Compounds from the organic nitrates group are relatively long-lived species and have been shown to have potential effects on atmospheric chemistry at local, regional and even global scales. In order to investigate the importance of these compounds in the indoor environment, Stephe R. Jackson et al. conducted experiments in a Teflon film environmental chamber to measure the formation of alkyl nitrates produced from α-pinene ozonolysis in the presence of NO and alkanes using gas chromatography with an electron capture detector (GC-ECD). Observed chemistry is similar to the one observed in atmospheric air. Concentration ratio of $[O_3]/[NO]$ around 1 was found to produce the highest organic nitrate concentration. With $[O_3]$ and $[NO]$ equal to 100 vppb and 105 vppb correspondingly, the organic nitrate formation was observed at a level of roughly 5 vppb. The experiments with α-pinene ozonolysis in the presence of NO suggest that organic nitrates have potential to form in indoor air via reaction of infiltrated ozone/nitric oxide and terpenes emitted from household products (Jackson et al., 2017).

Off-line measurement techniques were also successfully applied in real environment studies with real samples serving as a terpene emission source. The example of such study can be application of two dimensional gas chromatography coupled to high resolution mass spectrometry (GCxGC-TOF-MS) in studies on VOCs emission from herbs during pan frying. Application of this technique allowed to determine that frying meat with herbs and pepper caused emission of large amounts of mono-, di-, sesquiterpenes and terpenoids. The authors of this study emphasized that SOA formation via oxidation of terpenes emitted from herbs seems to be unfortunately overlooked in current indoor air quality control studies (Klein et al., 2016).

Other example of GC application in the studies on indoor SOA formation is the work of Toftum et al. (Toftum et al., 2008), who applied TD-GC–MS for determination of terpenoid substances emitted from wooden based products and SOA production potential via their ozonolysis. Four different types of wooden materials were investigated in this study: untreated pine shelf, Orientated Strand Boards, untreated beech boards and painted beech boards. Chromatographic analysis allowed to determine nine different terpenoid compounds and their concentration before and after ozone introduction into the chamber. Authors also managed to determine the importance of surface reactions of wooden materials, concluding that surface chemistry does not contribute significantly to the resulting SOA levels, however it does contribute to the ozone removal, which indicated that surface chemistry generates gas phase products, which should be analyzed in the future.

Gas chromatographic techniques are still widely used in broadly understood indoor air quality research, although it has its limitations. As an example, Toftum et al. (Toftum et al., 2008) described in their work, that one of the experiment with pine board was not successful due to the fact that sample was collected too quickly, while ozone concentration was still high, therefore determined concentration of terpenoids was very low probably due to their reaction with ozone. Application of real-time measurement techniques dedicated to gas phase analysis allows to omit such issues and observe concentration changes in a continuous way. Continuous collection of data facilitates determination of trace amounts of oxidation products and the description of the observed phenomena to a great extent.

In 2007 Nøjgaard et al. (Nøjgaard et al., 2007b) applied atmospheric sampling Townsend discharge ionization (ASTDI) source coupled to a triple quadrupole mass spectrometer (MS) for determination of oxidation products formed by limonene ozonolysis. The idea of this method is to sample directly from surrounding air (which allows to carry out on-line analysis),

ionize analytes by Townsend discharge source (which do not cause fragmentation of analyte) and finally to use MS for qualitative and quantitative analysis. This technique allows to differentiate between functional groups and to differentiate 1°, 2° and 3° alcohols, ketones, and aldehydes, which is an important advantage in case of terpene oxidation reactions. Main limitations of this proposed solution are complicated spectra and relatively low sensitivity in comparison to other real-time measurement techniques like Proton-Transfer-Reaction Mass Spectrometry (PTR-MS) (Nøjgaard et al., 2007a). In 2017 Liu et al. (Liu et al., 2017) successfully applied Vacuum Ultraviolet single-photon ionization time-of-flight mass spectrometry (VUV-SPI-TOF-MS) technique for analysis of volatile compounds and SOA formation initiated by stir-frying species, which are known source of terpenes indoors. VUV-SPI technique is a threshold ionization method based on the principle, that VOCs with ionization energy lower than single-photon energy will be ionized into molecular ions, which eliminates fragmentation. Due to the lack of chromatographic separation this technique is applied for on-line qualitative and quantitative VOCs analysis (Hou et al., 2007). Significant advantage of this technique is low detection limit of vppb (experiments done with benzene and SO_2) (Wu et al., 2010), or even sub vppb when a membrane enrichment device or heating device is combined (experiments done with benzene and chlorobenzene) (Yamamoto et al., 2010).

As popularity of real-time gas phase analysis techniques grew, more efforts have been put in their improvement. As a result, PTR-MS has been developed and introduced into the laboratory practice. The basis for PTR-MS development was Selected Ion Flow Tube (SIFT) technique developed in 1976 (Adams and Smith, 1976). Ion production was based on "afterglow" method, in which electronically excited gas constituents emitted very bright glow which caused ion generation. The emitted glow could reach from the source up to buffer gas region, hence the name "afterglow" (Blake et al., 2009). The application of a quadrupole filter allows to separate ions accordingly to their mass to charge ratio. Ions passing the quadrupole filter are directed into so called flow tube where they react with buffer gas (helium). Afterward, the mixture of ions in a buffer gas is directed to the second quadrupole analyzer and finally to the detector at the very end of the flow tube (Blake et al., 2009). Both PTR and SIFT techniques provide fast, easy, and direct analysis in real time, which is a massive advantage overcoming sample preparation and chromatographic separation steps in GC-based methods. They are both based on chemical ionization, followed by reaction in a specific reaction zone, followed by detection at the very end. Despite a lot of similarities those two techniques have two major differences. First difference is about the way of ion generation; SIFT-MS technique applies generation of wet air plasma by microwave discharge, followed by filtration of reagent ions (H_3O^+, NO^+ and O_2^+) by first quadrupole, whereas PTR-MS applies hollow cathode discharge that causes ionization of water to form H^3O^+ reagent ions. Second difference is about where reagent ions react with analyte; in SIFT-MS reaction takes place in a flow tube with an inert gas (helium or nitrogen), whereas in PTR-MS reaction takes place in a drift tube without the presence of an inner gas. Differences in those two methods are manifested by higher detection limits in SIFT-MS, higher risk of water clusters formation in PTR-MS and differences in fragmentation patterns for those two methods (Lehnert et al., 2019). PTR-MS is a very powerful instrument and very useful in VOCs monitoring and analysis, however its major drawback is that only nominal mass to charge ratio of the protonated compound can be determined. Therefore, in 2004 Blake and the rest of research team, for the first time successfully coupled PTR instrument with Time-of-Flight (TOF) analyzer (Blake et al., 2004). Implementation of TOF in a PTR-MS device allowed to differentiate between compounds of the same nominal mass to charge ratio and to detect trace gases at the vppb and sometimes even sub vppb level (Jordan et al., 2009). Today PTR-MS technique becomes more and more popular in various types of real time gas analysis. It is successfully applied in breath analysis (Moser et al., 2005; Herbig et al., 2009), atmospheric air research (Han et al., 2019; Li et al., 2020) and indoor air research (Ishizuka et al., 2010; Tang et al., 2015).

4.2. SOA particles characterization: particle number and mass concentration, particle size distribution

In general, particle measuring devices apply principle of light scattering to measure a range of particle features. For analysis of particle phase (Optical Particle Counter) OPC and (Condensation Particle Counter) CPC are most commonly applied (Klenø et al., 2006; Toftum et al., 2008; Coleman et al., 2008). OPC applies principle of light scattering to determine particle count and size distribution and CPC is considered to be an adjunct to OPC that extends the range of particle detection in a direction of smaller particle sizes by using condensation phenomenon before optical detection (Welker, 2012). Main limitation of CPC is that it can mainly measure total particle concentration, whereat OPC can only measure distribution of relatively large particles (0.3–10 μm aerodynamic diameter). Those features are considering to be a limitation, since currently, the most unknown process in SOA formation is nucleation at the beginning of the SOA formation. Nucleated clusters are characterized by very small diameter of few nm or less (Westervelt et al., 2013; Monod and Liu, 2012) therefore cannot be measured by OPC or CPC. Toftum et al. (Toftum et al., 2008) applied CPC (also known as CNC) working in the range 0.02–1 μm for measurement of particle number concentration, whereas OPC working in the range 0.1–2 μm was applied to determine particle mass concentration. In practice, it is very useful to measure both number and mass particle concentration. Sometimes there is no significant change in particle number concentration for the range of relatively large particles, while particle mass concentration in that region is changed, which suggests e.g., formation of small number of large particles. With only number concentration measured, one could suggest that large particles were not formed at all (Morawska et al., 1999). Another example of the application of CPC is the study dedicated to assessment of effectiveness of plants (11 different species of potted plant, shrubs and trees) to decrease ultrafine particle concentrations in indoor environments. It has been investigated that most of the plants showed typically small but statistically significant activity in particle concentration reduction. Study results proved that there is a linear relation between number of plants and number of particles reduced and also linear relation between plant leaf area and number of particles reduced (Stapleton and Ruiz-Rudolph, 2018).

Other instruments successfully tested for particle phase measurements are aerosol monitors (e.g.: OPC, CPC, P-Track, LASAIR). They allow to measure size and concentration of suspended particles in real time; however, the device can measure particles of relatively large diameter 0.1 μm to 15 μm. Due to the range of measurement and small size of the device it is mainly used for fast field measurement to determine PM1, PM2.5 and PM10 (Cincinelli et al., 2016; Khurshid et al., 2016). Aerosol monitor was successfully applied by Khurshid et al. (Khurshid et al., 2016) to determine particulate ROS present in indoor environment, proving that about 58 % of particulate ROS was present on PM2.5. ROS importance in indoor environment will be described in more details in further part of this paper. Apart from OPC and CPC, scientists also applied a range of other particle counters that differ with operation principle and range of particle size to be measured. Among those, a 6-channel handheld particle counter able to measure particles of diameter > 300 nm is very useful in case of field measurements. Around year 2000 scientists started to suggest that certain health effects may be more related to particle number concentration than to particle mass concentration (Donaldson et al., 1998) (Johnston et al., 2000). Therefore a simple 6-channel counters began being substituted by ultrafine particle counters that allowed to measure ultrafine particles of diameter 1–100 nm, which is much more useful in case of investigation of SOA formation (Yu et al., 2011; Stapleton and Ruiz-Rudolph, 2018). Sometimes two particle counters were applied together to ensure wider particle size range to be measured. For example Sarwar et al. (Sarwar and Corsi, 2007) applied P-TRAK and Lasair® particle counters in their experiments aimed for determination of limonene/ozone reaction impact on indoor SOA formation, however application of those two techniques allowed to investigate only particles of size in the range 0.02–42 μm. Authors indicated in their work, that it is necessary to investigate the reaction of SOA

formation initiated by limonene/ozone reaction at the initial phase of the experiment with the application of techniques that allow to determine finer particles (with single nm in diameter) (Sarwar and Corsi, 2007). Due to the need of investigation of nucleation process, optical counters became supported by newly developed techniques for particle size differentiation based on particle electrical mobility such as Scanning Mobility Particle Sizer (SMPS). The fact that CPC can measure only particle concentration and cannot provide information about particle sizes caused development of SMPS devices. SMPS principle of operation relies on detection with CPC and additionally implementation of Differential Mobility Analyzer (DMA) before CPC detection. It allows to differentiate between particles of different sizes. At the inlet of SMPS, particles are firstly selected by an impactor, then neutralized to minimize the risk of wall losses, further particles are differentiated accordingly to their size in a DMA, and finally detected and counted by CPC. Application of DMAs of different length allows to measure different size ranges of particles. The "long" DMA allows to measure particles of 10–1000 nm (TSI Incorporation, 2021b), whereas "nano" DMA allows to measure particles of 2–165 nm (TSI Incorporation, 2021a). Particle counters, which principle of operation is based on differences in particles electrical mobility, are widely used in indoor SOA particle number and size distribution studies. Such spectrometer has been for instance applied for steady state chamber experiments to measure the SOA formation entirely initiated by ozone reactions with squalene sorbed to glass. Squalene as a non-volatile compound tends to adsorb on indoor surfaces. It has been proved that on-surface squalene ozonolysis forms significant amounts of SOA indoors, even at low ozone and low humidity conditions (Wang and Waring, 2014).

Ultrafine particle concentration in air is rapidly changing, therefore sometimes SMPS works too slow by typically taking 30 s or longer to measure particle size and distribution. Therefore, recently scientists from Tartu University developed a device based on electrical aerosol spectrometer technology – Fast Mobility Particle Sizer (FMPS). FMPS can measure particle size from the range 6–560 nm with 16 channels per decade every second which makes it faster than SMPS. However, there are still some doubts about precision and reliability of FMPS comparing to SMPS. Studies revealed some significant discrepancies concerning both: measurement of particle size and measurement of particle concentration. Research carried out in 2009 proved that (i) particle number concentration measured by SMPS is lower than particle number measured by FMPS due to diffusion loses in SMPS and application of diffusion correction factor is needed, (ii) particle size distribution measured by SMPS is different than that measured by FMPS, and application of correction factor is needed. To improve FMPS size distribution, it can be corrected by comparing FMPS distribution data with DMA-segregated particle counts measured by CPC (Jeong and Evans, 2009). Another study from 2013, where FMPS was compared with SMPS and HR-TOF-AMS revealed that FMPS underestimates measured particle sizes (by 40–50 %). Because FMPS by default is delivered without any calibration tool, it is important to correct particle size by employing size-calibrated DMA to produce monodisperse particles to be sampled by FMPS (Lee et al., 2013).

Brief description of advantages and limitations of strategies applied in indoor air characterization regarding both chemical composition of gas phase and particle size and concentration distribution is presented in supplementary Table S1.

4.3. Characterization of reactive oxygen species (ROS) as a significant factor in SOA formation and transformations

At the beginning of 2000 scientific interest focused on ROS that are formed via terpene oxidation reactions and play a significant role in SOA chemistry. ROS include molecules such as organic peroxides and hydroperoxides, some ions (e.g., hypochlorite ion, peroxynitrate ion) and radicals (e.g., hydroxyl, alkyl peroxyl). According to literature data, ROS can be formed via photochemical reactions (with NO_x, CO, formaldehyde or VOCs) (Gunz and Hoffmann, 1990) and via ozone initiated reactions (Weschler, 2006). Terpene and ozone reactions have been suggested to be

an important pathway of indoor H_2O_2 and organic peroxides species formation, under conditions of minimal photochemical activity (Fan et al., 2005; Docherty et al., 2005). It has been proved that indoor concentration of limonene and ozone strongly influence ROS concentration and that major fraction of indoor ROS exist on PM2.5 (Khurshid et al., 2016). ROS in air can occur freely in (i) gas phase, (depending on their oxidation degree) (ii) adsorbed onto existing particles, (iii) and/or ROS can nucleate to form new particles. Gas phase ROS are expected to absorb in the mucus of upper airways and be removed from respiratory track, while particulate ROS can be transported deeper into respiratory track, where they come into direct contact with lung tissue and further into the bloodstream reaching other organs (Bailey et al., 1985; Semmler et al., 2004). Therefore, pulmonary and systematic oxidative stress caused by ROS is stated to be important molecular mechanism of particulate matter-mediated toxicity (Gurgueira et al., 2002; Kehrer, 1993). The biggest struggle with ROS investigation is the time between sampling and analysis. Mostly, samples are collected and refrigerated until analysis due to difficulties in carrying out in situ measurement. Time interval between sampling and analysis may allow samples to decay or undergo re-equilibrium, which causes bias that results in underestimation of ROS concentration (Chen and Hopke, 2009b; Chen et al., 2011). In 2010 Chen et al. (Chen and Hopke, 2010) investigated SOA formation via limonene and ozone reaction. In this study authors additionally investigated how the time between ROS sampling and analysis influences result. ROS were determined using fluorogenic probe, dichlorofluorescein (DCFH) (Venkatachari et al., 2005; Hung and Wang, 2001), which is still the most commonly applied method for ROS determination (Khurshid et al., 2016; Chen et al., 2017). The idea of the method is that DCFH is a non-fluorescent reagent until it becomes fluorescent due to reaction with ROS species. This reaction needs a catalyst which most commonly is horseradish peroxidase (HRP) that is added to the DCFH solution. All preparation and analysis must be done in dark, only with red light due to photo sensitivity of DCFH-HRP working solution. Finally, fluorescent intensities must be converted for to equivalent H_2O_2 concentration by conducting an assay of standard calibration. Detailed description of the method can be found elsewhere (Venkatachari et al., 2005; Chen and Hopke, 2009a). Chen et al. (Chen and Hopke, 2010) analyzed ROS samples after 5 min after sample collection, samples refrigerated at −20 °C and samples kept at room temperature for 24 h before analysis. Loss of ROS between samples analyzed immediately and after 24 h were manifested with approximately threefold loses of signal for 24 h storage samples. It is due to ROS reactions with some other collected species during storage time. Authors also noticed difference between reactivity and volatility between samples stored in the freezer and stored at room temperature. One important information about Chen et al. (Chen and Hopke, 2010) experiments is that sampling time was equal to 30 min, while there are some ROS with lifetime shorter than sampling period (30 min) which are lost due to the sampling procedure. In 2016 Wragg et al. (Wragg et al., 2016) came up with a new portable Online-Particle-Bound ROS Instrument (OPROSI) that allows for fast quantification of particle-bound ROS. OPROSI is based on constant sample inflow through the system, which prevents from ROS losses due to sampling and storage, whereas sample analysis and concentration determination is performed using fluorescent DCHF-HR method. Detailed description of the OPROSI method is presented elsewhere (Wragg et al., 2016). In 2017 OPROSI was successfully applied by Gallimore et al. (Gallimore et al., 2017a) in chamber studies aimed at analysis of ROS formation initiated by limonene oxidation in relation to indoor air. This method allowed to determine two categories of ROS formed via limonene and ozone reaction. Based on time series analysis authors identified short-lived components produced during precursor ozonolysis, characterized by lifetime of the order of minutes and stable, long-lived components (4 h) (Gallimore et al., 2017a). This study indicates importance of analysis performed at the very beginning of the terpene oxidation reactions, because short-lived ROS components can react within seconds or minutes after sampling. Therefore, main goal in ROS investigation is to develop analytical procedure that will minimize the time between sample collection and analysis. On-line tandem MS appeared as a promising tool in this case (Tomaz et al., 2021). In the referenced study, several structures that could be potential radicals formed during limonene autooxidation and peroxy acid functional groups in limonene and α-pinene peroxy radicals have been determined.

4.4. Condensed phase analysis

There is a lot of studies focused on investigation of SOA particle size and mass distribution, however chemical composition of aerosol is extremely important, especially while evaluating toxicity of the aerosol.

Due to the fact that there are still some discrepancies between laboratory experiments and models evaluating SOA formation, it is necessary to provide as much data about SOA formation to the model as possible (Kroll and Seinfeld, 2008; Westervelt et al., 2013; Monod and Liu, 2012). Over the past 20 years a lot of analytical techniques have been developed for determination of SOA chemical composition. Those methods can be divided into on-line and off-line. Among the on-line techniques it is common to apply Aerosol Mass Spectrometry (AMS) to explore chemical SOA composition with high time resolution (Cross et al., 2007; Sullivan and Prather, 2005). AMS allows to determine particulate organic matter nature – sulfate, nitrate, ammonium and chlorine content and oxidation state with functional group identification. Wang et al. (Wang et al., 2019) applied AMS to study SOA formed via most common indoor VOCs arising from use of cleaning products, fragrance, and air fresheners, with HOCl and Cl_2. Presence of HOCl and Cl_2 in the indoor air results in high chlorine species contribution in resulting SOA and formation of high molecular weight products. Two sets of experiments were conducted in the absence and presence of day light, as light is essential factor in formation of particles from limonene oxidation by HOCl and Cl_2. Sunlight diffused by window provides enough energy to cause photolysis of HOCl and Cl_2 leading to formation of Cl and OH radicals and ultrafine particles. AMS was applied to determine chemical composition of particles with vacuum aerodynamic diameter 70–100 nm. Main goal was to identify and monitor the relative temporal trends of different classes of chemicals in the aerosol rather than to provide a quantitative analysis of aerosol mass. However, AMS is characterized by hard ionization source that causes fragmentation of chemical compounds, therefore, off-line techniques are also frequently applied (Wang et al., 2019).

Off-line techniques differ between an on-line ones mainly by the sample preparation step which is omitted in on-line ones. Sample collection and extraction enable to access the high complexity of SOA composition (Rossignol et al., 2012). Most commonly applied procedure of off-line techniques involves aerosol sample collection on filters (e.g. PTFE) followed by extraction with a solvent (most commonly methanol), sonication and further analysis. Hu et al. (Hu et al., 2017) in their study applied collection of aerosols on filters, which was followed by extraction and analysis with FTIR. The main goal of their study was to investigate NO_2 influence on SOA formation initiated via limonene ozonolysis. Application of an off-line FTIR technique allowed discovering 385 condensable compounds characterized by high influence on SOA mass (93–96 %). Among of all discovered compounds 20 were with >7 carbon (C) atoms. Results presented by the authors confirming the thesis that compounds with more than 6C atoms have the major potential of SOA formation (Seinfeld and Pandis, 2016; Hu et al., 2017). In previously mentioned study of Wang et al. (Wang et al., 2019), in order to provide more molecular data to the results obtained by on-line AMS, off-line ESI-MS technique was also applied. It allowed for determination of high molecular compounds like monomers, dimers, and trimers of limonene oxidation products (up to 900 m/z).

Off-line procedures are constantly manipulated and developed, e.g. Riva et al. (Riva et al., 2016) investigated OH scavengers influence on the SOA formation from isoprene ozonolysis. For SOA chemical composition authors applied two procedures: one for UPLC/ESI-HR-TOFMS and second for GC-EI/MS analysis. The beginning of the procedure in case of above mentioned techniques was the same, starting with collection of aerosols on filters, followed by extraction with methanol, sonication, drying methanol extracts with nitrogen, reconstitution in methanol and then division of

extracts to perform further analysis using previously mentioned two instrumentation sets. For UPLC/ESI-HR-Q-TOFMS set samples were again blow dried under stream of N_2, again reconstituted in MeOH mixture with purified water and analyzed. For GC-EI/MS analysis, filter extracts were trimethylsilylated by addition of BSTFA, trimethylchlorosilane and pyridine mixture, then heated and analyzed within 24 h from trimethylsilylation. Chemical characterization of SOA showed significant yields of OSs and 2-methyltetrols from ozonolysis of isoprene in the presence of acidified sulphate aerosol. The presence of an OH radical scavenger in chamber studies resulted in a reduction in the concentration of products that could be identified, indicating the involvement of OH radicals in oxidation. Authors indicate that substantial particle phase concentration of organoslufates and 2-methyltetrols from ozonolysis in the presence of OH scavenger suggest that SOA yields from isoprene ozonolysis may be underestimated by current models and that contribution of ozonolysis to the isoprene-derived SOA budget is underestimated (Riva et al., 2016). Generally, extraction of sample provides a huge range of possibilities of further studies. However, Wang et al. (Wang et al., 2019) have noticed that species collected on the filters can react with filter substrate, semivolatiles may evaporate from filter surface during collection, which can result in increased concentration of oligomers and decreased concentration of volatiles at the end of measurement. Therefore, there is still a need for further development of SOA chemical composition investigation methods.

5. Highly oxygenated molecules as a new pathway of indoor SOA formation

Highly Oxygenated Molecules (HOM) were recently discovered and are concerned as new pathway for SOA formation via VOC oxidation (INDAIRPOLLNET, 2020). It has been proved that monoterpenes are able to rapidly generate HOMs via autooxidation process (Bianchi et al., 2019). Since the term HOM is relatively new, some problems with its definition occurred. Therefore Bianchi et al. in 2019 came out with a review article in which authors try to define more precisely what HOMs refers to. They proposed a guideline for classification of compounds as HOMs, which are as follows:

- HOMs are formed via autooxidation involving peroxy radicals.
- HOMs are formed in the gas phase under atmospherically relevant conditions.
- HOMs typically contain six or more oxygen atoms in the structure.

Bianchi et al. (Bianchi et al., 2019) team also analyzed relationship between HOMs and already well-known volatility classes of VOCs. According to the review one can distinguish 5 volatility classes:

- Extremely Low Volatility Organic compounds (ELVOC)
- Low Volatility Organic Compounds (LVOC)
- Semivolatile Organic Compounds (SVOC)
- Intermediate Volatility Organic Compounds (IVOC)
- Volatile Organic compounds (VOC)

Deep analysis of properties characteristic for each volatility group revealed that HOMs mainly fall into ELVOC group, which describes molecules that condense onto any pre-existing cluster and may also participate directly in new-particle formation involving only other ELVOC (Bianchi et al., 2019) and that ELVOCs are formed within first few minutes after oxidant addition (Jokinen et al., 2015).

Autooxidation of VOCs is an oxidation of hydrocarbons by atmospheric oxygen. This reaction in liquid phase is well-known since 1875 (Jazukowitsch, 1875). Autooxidation in gas phase was unimportant for atmospheric VOC oxidation, because chain propagation via intermolecular H-abstraction is unlikely to happen in the atmosphere due to relatively low hydrocarbon concentration (few vppb) (Jokinen et al., 2014). However, studies published in 2012 and 2014 aimed at investigation of

formation of ELVOCs in boreal forest and in chamber studies with α-pinene oxidation prove that monoterpene oxidation leads to formation of HOMs and that oxidation process leading to HOM formation most likely takes place in the gas phase (Ehn et al., 2012; Ehn et al., 2014). VOCs oxidation leads to formation of highly oxidized RO_2 radicals, and further closed-shell products are expected to have extremely low volatility, which makes them crucial in SOA formation in atmosphere (Jokinen et al., 2014). It has been also reported that compound containing endocyclic double bond efficiently produce HOMs under atmospherically relevant concentrations whereas HOM formation initiated by ozonolysis is higher than from reaction with OH radical. However OH radical initiated HOM formation increases while the compound has exocyclic double bond or when it's acyclic (Jokinen et al., 2015).

HOMs were firstly observed in 2014 and described in paper about field studies in boreal forests, and then those results were confirmed by laboratory chamber studies of monoterpene oxidation (Ehn et al., 2012; Ehn et al., 2014; Ehn et al., 2010; Zha et al., 2018; Bianchi et al., 2017; Wang et al., 2017; Molteni et al., 2018).

The ability of HOM detection emerged from recent advances in mass spectrometry with atomic pressure interface time-of-flight mass spectrometry (APi-TOF-MS) that is applied for HOM detection alone or coupled with nitrate ion chemical ionization source (NO_3^- Cl-APi-TOF-MS). Application of this analytical technique allowed to determine HOMs both in gas and particle phase (INDAIRPOLLNET, 2020; Kürten et al., 2016; Ehn et al., 2012; Ehn et al., 2014; Berndt et al., 2016, 2018; Molteni et al., 2018; Ehn et al., 2010; Bianchi et al., 2017; Zha et al., 2018). API-TOF-MS allows to sample ions directly from ambient air due to combination of atmospheric pressure interface unit directly connected to time-of-flight spectrometer. For sampling neutral molecules APi-TOF-MS must be equipped with a chemical ionization source, then neutral molecules are artificially created before reaching APi-TOF (Jokinen et al., 2012). Chemical ionization is achieved by exposing clean air containing HNO_3 to alpha radiation or X-rays, which results in formation of nitrate ions. Nitrate ions are later directed, by electrical field, into the sample flow. When nitrate ions reach the sample, neutral molecules become charged due to adduct formation (e.g., with HOM) or by proton transfer reaction (e.g., with sulfuric and some dicarboxylic acids). Ionized molecules are then directed to the TOF analyzer (Bianchi et al., 2017). Currently, there is no direct calibration for HOMs, and methodology described by Ehn et al. (Ehn et al., 2014) involving calibration constant for sulfuric acid is applied to determine HOM concentration (Bianchi et al., 2017).

Although there was quite a lot of studies focused on investigation of HOMs formation and their role in SOA formation in atmospheric air, little was done about investigation of HOMs chemistry in indoor air. There is one outstanding paper of Pagonis et al. (Pagonis et al., 2019), where autooxidation of limonene in the museum is investigated. Experiments were performed at University of Colorado Art Museum for over 6 weeks in 2017. VOCs concentration was monitored using PTR-Q-MS, whereas HOMs were investigated using high-resolution time-of-flight nitrate adduct chemical ionization mass spectrometer (NO_3^- CIMS). Experiment with peeling an orange inside the museum showed significant increase in concertation of following HOMs after the peeling activity: $C_9H_{14}O_5$, $C_7H_{11}NO_7$, $C_9H_{14}O_7$, $C_{10}H_{17}O_7$, $C_9H_{13}O_9$ and $C_{10}H_{15}NO_9$. Nitrogen present in some of the HOMs could emerge from limonene reaction with NO_3 radical, however, those reactions made negligible contributions to HOM formation. It was also determined, that increasing limonene concentration is followed by parallelly increasing HOM concentration and most of the HOMs are produced due to monoterpene ozonolysis reaction. Since it is well-known that activity such as peeling an orange indoors causes SOA formation increase, authors investigated also impact of HOMs formation on aerosol mass in the museum and concluded that HOMs significantly influence SOA yield indoors. Although outdoor studies on how HOMs influence SOA formation can be applicable for indoor air in some cases (few tens vppb of ozone and low concentration of NO), those two environments are different and investigation of HOMs chemistry indoors is still needed (Pagonis et al., 2019). Other study based on the application of on-line tandem MS

revealed that HOM dimer formation during limonene ozonolysis is driven by O_8 peroxy radical, whereas O_4 peroxy radical seems to be a key component for dimer formation during α-pinene ozonolysis (Tomaz et al., 2021).

Selected literature examples of chamber and real sample/real environment studies dedicated to gaseous components determination, SOA particles characterization, SOA chemical composition determination, HOM characterization and ROS characterization relevant to SOA formation in indoor air are presented in supplementary Tables S2 and S3 respectively. The tables summarize information on applied measurement techniques, measured parameters, individuals and compounds as well as the obtained results, which in many cases relate to the chemical analysis of the test sample. Understanding SOA in indoor air is not only about measuring the number of particles, their size or distribution, but also the chemical composition of the particles, as this aspect determines changes in the composition of the particles themselves. In most cases these are model studies, there are only a few examples of analysis of real samples where the chemical composition and particle distribution are studied in parallel. Modeling studies are necessary and essential but they can never duplicate studies where real samples are analyzed. Therefore, there is a continuous need to broaden the scope of real environment studies, which would aim not only at SOA particle characterization but also at detailed SOA chemical composition determination.

6. Summary, conclusions and future perspectives

The main pathway of SOA formation indoors is via oxidation of reactive VOCs present in indoor air (Youssefi and Waring, 2014; Niu et al., 2017). Terpenes are reactive VOCs present in indoor air due to wide range of emission sources such as household cleaning and disinfectant products (Carslaw, 2013), building materials, furniture (especially wooden) (Missia et al., 2010), cosmetic products (Tsigonia et al., 2010), herbs and spices (Klein et al., 2016), additionally human beings serve as a source of reactive VOCs, like squalene, themselves (Xiong et al., 2019; Yang et al., 2021a; Yao and Zhao, 2021). In the indoor environment, new particle formation (NPF) initiated by reactive VOCs oxidation may occur due to nucleation process or due to gas-to-particle partitioning. Those processes strongly influence particle size distribution in a range of ultrafine (<0.1 μm) particles (Michael S. Waring and Siegel, 2013). Most of the studies focus on gas-phase reactions of terpenes and/or terpenoids with oxidants (most commonly ozone and hydroxyl radicals) leading to NPF indoors (see Tables S2 and S3 for examples). However, there are literature data proving that surface reactions are also relevant regarding NPF indoors, since indoor surfaces are complex mixtures of films containing a range of reactive VOCs (Liu et al., 2003; Weschler and Nazaroff, 2008). Moreover, terpenoids, due to their vapour pressure, exerts moderate sorption onto indoor surfaces from the gas phase (Corsi, 2001) and they also can get onto the surfaces directly from using household products (Nazaroff and Weschler, 2004). As the primary ozone loss mechanism indoors is via deposition on surfaces (Sabersky et al., 1973), the terpene/terpenoid ozonolysis reaction takes place on household surfaces too (Waring and Siegel, 2013).

Indoor SOA new particle formation (NPF) events and further size distribution evolution depends on indoor environmental conditions, such as ventilation, air cleaning/filtration, high surface are-to-volume ratios, presence of surfaces that may serve as sinks due to SOA deposition, sources or media for heterogeneous chemistry for particles and reactive gases, nonphotolytic radical sources (such as ozone – alkene reactions) (Youssefi and Waring, 2014; Niu et al., 2017), but also temperature, humidity and initial substrate concentration (Jonsson et al., 2008). There is a vast number of studies on different environmental factors influencing SOA formation, thus we would like to stress some the most important findings. There are literature findings proving that increase air exchange rate decreases aerosol mass fraction (also called SOA yield) (Youssefi and Waring, 2014). Increased limonene – ozone ratio increases aerosol mass fraction (Youssefi and Waring, 2014; Rösch et al., 2017) Application of ionization air purifiers may actually increase the mass concentration of fine and ultrafine SOA, when unsaturated VOCs (like limonene) are present (Alshawa et al., 2007). Surface oxidation reactions promote nucleation process even more than gas-

phase reactions (Waring and Siegel, 2013). Most of the studies confirm negative or no effect of increased RH on SOA number concentration (Hessberg et al., 2009; Wang and Waring, 2014) and positive effect on SOA mass concentration (Jonsson et al., 2006; Chen et al., 2021b). However, the results are not consistent and some studies indicate the opposite results (Jonsson et al., 2006; Tillmann et al., 2010). It was also found that SOA yield (Hoffmann et al., 1997) and aerosol formation potential are higher at lower temperatures due to more active condensation of organic vapors under lower temperatures (Vu et al., 2013). However, Jonsson et al. claimed in their study that the temperature and SOA formation dependence is not so clear and even a positive effect was observed that has to be related to changes in chemical mechanism and/or reaction rates at low temperatures (Jonsson et al., 2008). One of the biggest struggles in this study area is that the reactions of oxidation of reactive organic compounds and formation of oxidation products and aerosol in indoor air are very fast, occurring in a comparable time or faster than air exchange rate. Recently, it has also been discovered that SOA formation may be enhanced by rapid formation of highly oxygenated organic molecules (HOMs), also called extremely low volatility organic compounds (ELVOCs), which is yet another factor that would have to be incorporated while considering indoor air chemistry.

All the above-mentioned factors influencing SOA yield, particle number and mass concentration and particle size distribution makes the research in terpene/terpenoid initiated SOA formation subject extremely difficult task. Therefore, a lot of research is done by using chambers (environmental/reaction chambers). Mimicking indoor conditions in the chamber facilitates research due to possibility of control the amount and intensity of factors influencing terpene/terpenoid oxidation initiated SOA formation. However, according to Tables S2 and S3, research in this field of study are conducted using different types and sizes of chambers, either in static or dynamic conditions, under light (different intensities and sources) or dark conditions, using different substrates and different initial substrates concentrations, with presence/absence of seed aerosol, with presence/absence of OH scavengers, applying different observation times, under different temperature and RH, using different analytical approaches (either real-time measurement techniques or off-line techniques). It is known that research vary in terms of its purpose and scope, additionally, in most cases it is also difficult to compare studies – even with the same or similar aim - due to differences in a way of conducting the experiment.

Due to the struggles with reactive VOCs oxidation chemistry analysis and SOA formation analysis, a range of analytical techniques was developed over the last years to help understand the complicated chemistry in indoor and atmospheric air. A great analytical progress in SOA characterization was made by development of SMPS that allows to analyse SOA formation at the very beginning of the process. However, still, according to the data presented in Tables S2 and S3, most of the research regarding indoor SOA formation/generation, that were published during recent years are focused on particles with diameter >5.5 nm (mostly 5.5–800 nm). There is an observable gap in knowledge regarding formation of particles with diameter smaller than 5 nm and that's the range in which nucleation processes of stable nuclei, the first step of SOA formation, takes place. One of the most important papers recently published, concerning particles of diameter smaller than 5 nm is paper by Yang et al. from 2021. The study for the first time reports nanocluster aerosols (NCA, particles <3 nm) formation via ozone reaction with human skin surface. Authors report that ozone initiated chemistry with human skin lipids cause formation of particles with diameters in a range 1.18–1.55 nm and these nanoparticles are precursors for NPF, which is a key phenomenon associated with the development of indoor aerosol (Yang et al., 2021b).

There is a lot of research concerning SOA formation via oxidation of reactive organic compounds in atmospheric air, however this knowledge can only be partially applied for indoor air due to significant differences between those two environments. Increasing interest in indoor air quality regarding indoor aerosol monitoring and SOA formation is clearly visible by increasing number of scientific papers regarding this subject published during las decade, depicted in supplementary Fig. S1. Current pandemic

situation for sure increased the importance of indoor air quality monitoring and indoor air chemistry research. Even though analytical techniques evolved a lot from simple detectors and basic gas chromatographic techniques to real-time measurement techniques, which significantly expand the area of acquired knowledge, there still are gaps in information regarding details of SOA formation processes in indoor air.

Vast number of studies indicate negative effect of terpene induced SOA on living organisms, such as upper airway irritation, eye irritation and inflammatory response in the cells (Lin et al., 2016; Wolkoff et al., 2000; Clausen et al., 2001; Wolkoff et al., 2013). It is also proven that inhalation of aerosol is linked to worsening asthma, cardiovascular diseases, hypertension, diabetes, cancer lesions, central nervous system dysfunction as well as low birthweight (as a consequence of pregnant woman exposure) (Schraufnagel, 2020). Currently, air quality standards set limit values only for larger particles, i.e. PM2.5 and PM10, but there are no guidelines for ultrafine particles, which may affect human organisms in a different way than larger fractions (Marval and Tronville, 2022). The smaller the particles, the more toxic they can be for human health due to the increasing surface-to-volume ratio with the lowering particle size. Consequently, the surface of smaller particles potentially represents a much more extensive interface to transmit toxic chemicals than in the case of larger particles (Marval and Tronville, 2022). It is also proven that suspended particles may contain and transfer in indoor air bacteria, viruses, fungi and/or their metabolites as well as plant pollen and fragments of plant tissues (Domingo et al., 2020). The specific health effects of ultrafine particles remain unknown, but exposure to higher concentrations of nanoparticles has been associated with adverse health effects due to their deep penetration into the human lung and even transport to the brain via neurons (Marval and Tronville, 2022). Additionally, the fate of inhaled SOA is strongly size-dependent (Manigrasso et al., 2018; Guo et al., 2020).

We would like to highlight, that despite the issues and gaps in knowledge described in this review, there are still upcoming new research articles indicating new findings and new areas requiring further insight. As an example, Chen et al. in 2021 described differences in reactivity between the peroxy radicals from limonene oxidized by OH, NO3, and Cl oxidation claiming that NO_3^- and Cl^- derived peroxy radicals have faster rate coefficients than the ones derived from OH^-. Authors also indicated that in environments with low concentrations of HO_2 and NO, efficient autoxidation will lead to the formation of highly oxygenated organic compounds and thus likely aid in the growth of SOA (Chen et al., 2021a). Last year's COVID-19 pandemic caused increase usage of disinfectant agents; Rosales et al. in their 2022 research indicated that cleaning activities increased indoor monoterpene concentration by two orders of magnitude in comparison to the outdoor concentration and that increase enhanced ozonolysis reaction even under low ozone level (<10 ppbv). This, in turn, resulted in burst of ultrafine particles in such high concentration that respiratory tract deposited dose rates were comparable to or exceeding those of inhalation of vehicle associated aerosols (Rosales et al., 2022). Apart from the studies concerning every-day life at homes, schools, or offices, one cannot neglect the specific kinds of environments that are prone to high reactive VOCs concentrations. In 2021 we have published the study focused on terpene concentration in SPA salons We have found that excessive usage of essential oils greatly increases indoor terpene concentration, thus delivering SOA precursors (Pytel et al., 2021). Recently, in 2022 Kaikiti et al. conducted similar studies in hairdresser salons, indicating that application of hair products significantly increases reactive VOCs concentration in the salons and as a result measuring levels for PM revealed violations of the EPA and WHO international standards for permissible limit concentrations (Kaikiti et al., 2022).

Real indoor environment especially requires further characteristics, since currently chamber studies outweigh the field studies, and it is well known that chamber studies do not reflect real environment in 100 %. We are still struggling with precise mechanisms descriptions, uniform experimental approach, and application of our knowledge to different types of indoor environments.

CRediT authorship contribution statement

Klaudia Pytel: Visualization, Writing – original draft. **Renata Marcinkowska:** Writing – original draft. **Małgorzata Rutkowska:** Visualization, Writing – original draft. **Bożena Zabiegała:** Conceptualization, Supervision, 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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.156948.

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