

Journal Pre-proof

Biogenic secondary organic aerosols: A review on formation mechanism, analytical challenges and environmental impacts

Mithlesh Mahilang, Manas Kanti Deb, Shamsh Pervez



PII: S0045-6535(20)31966-4

DOI: <https://doi.org/10.1016/j.chemosphere.2020.127771>

Reference: CHEM 127771

To appear in: *ECSN*

Received Date: 9 May 2020

Revised Date: 15 July 2020

Accepted Date: 19 July 2020

Please cite this article as: Mahilang, M., Deb, M.K., Pervez, S., Biogenic secondary organic aerosols: A review on formation mechanism, analytical challenges and environmental impacts, *Chemosphere*, <https://doi.org/10.1016/j.chemosphere.2020.127771>.

This is a PDF file of an article that has undergone enhancements after acceptance, such as the addition of a cover page and metadata, and formatting for readability, but it is not yet the definitive version of record. This version will undergo additional copyediting, typesetting and review before it is published in its final form, but we are providing this version to give early visibility of the article. Please note that, during the production process, errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

© 2020 Elsevier Ltd. All rights reserved.

**Biogenic secondary organic aerosols: A review on formation mechanism,
analytical challenges and environmental impacts**

Mithlesh Mahilang, Manas Kanti Deb*, Shamsh Pervez

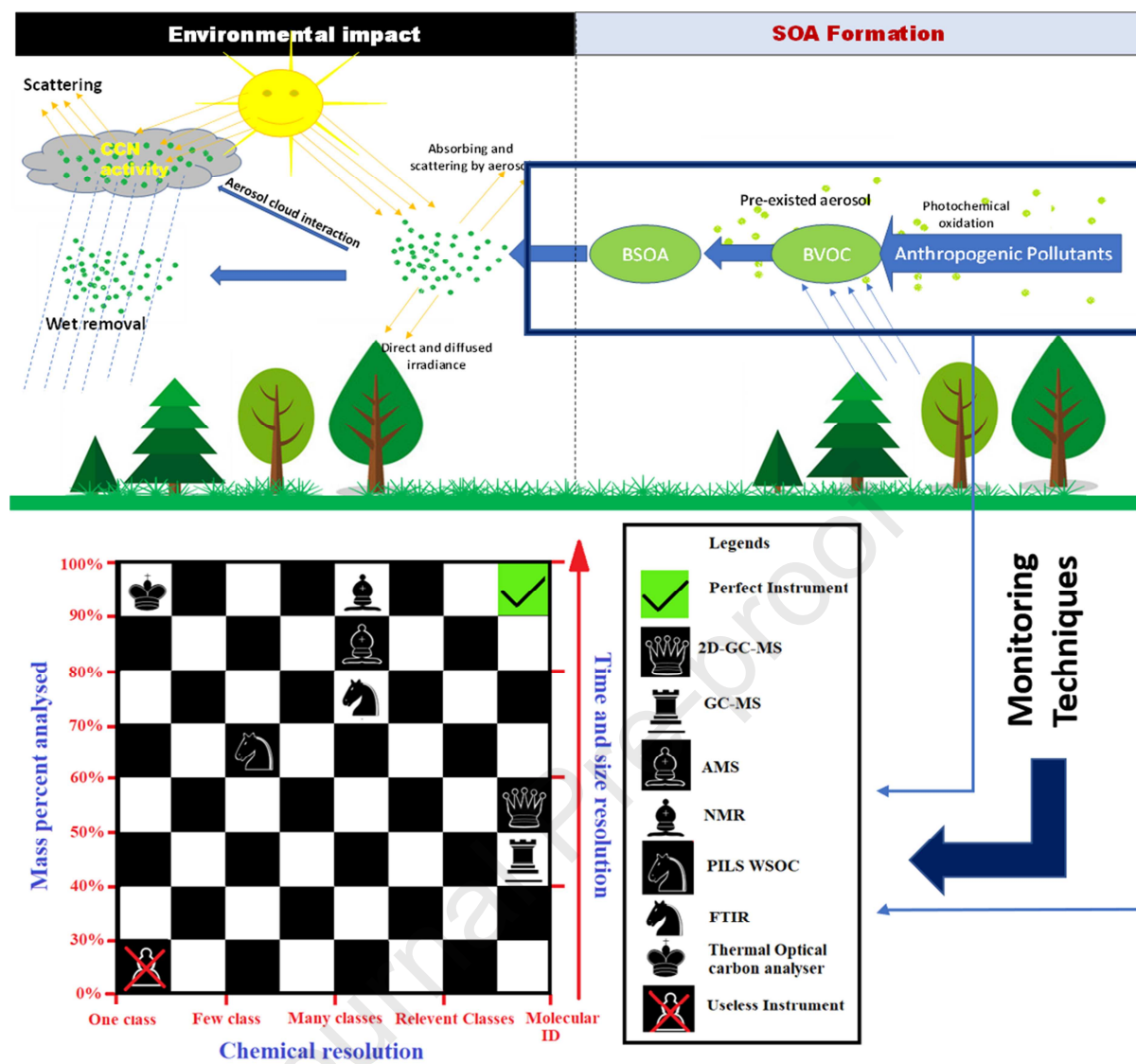
School of Studies in Chemistry, Pandit Ravishankar Shukla University, Raipur, Chhattisgarh, 492010,

INDIA

*Corresponding Author

E-mail: debmanas@yahoo.com (Dr. Manas Kanti Deb)

Contact: +91 9425503750, Fax: 91(771)226258.



Graphical abstract

**Biogenic secondary organic aerosols: A review on formation mechanism,
analytical challenges and environmental impacts**

Mithlesh Mahilang, Manas Kanti Deb*, Shamsh Pervez

School of Studies in Chemistry, Pandit Ravishankar Shukla University, Raipur, Chhattisgarh, 492010,
INDIA

*Corresponding Author

E-mail: debmanas@yahoo.com (Dr. Manas Kanti Deb)

Contact: +91 9425503750, Fax: 91(771)226258.

Abstract

The review initiates with current state of information on the atmospheric reaction mechanism of biogenic volatile organic compounds (BVOCs) and its fate in the atmosphere. The plants release BVOCs, i.e., isoprene, monoterpenes, and sesquiterpenes, which form secondary organic aerosols (SOA) upon oxidation. These oxidation reactions are primarily influenced by solar radiations along with other meteorological parameters viz.; temperature and relative humidity, therefore, the chemistry behind SOA formation is different during day than the night time. The review throws light upon the day and nighttime formation mechanism of SOA, recent advancements in the analytical techniques available for the measurements, and its impact on the environment. Studies have revealed that day time SOA formation is dominated by OH and O₃, however, NO_x initiated SOA production is dominated during night. The formation mechanism addresses that the gaseous products of VOCs are firstly formed and then partitioned over the pre-existing particles. New particle formation and biomass-derived aerosols are found to be responsible for enhanced SOA formation. 2-Dimensional gas chromatography-mass spectrometer (2D-GC/MS) is observed to be best for the analysis of organic aerosols. Radiative forcing (RF) SOA is observed to be a useful parameter to evaluate the environmental impacts of SOA and reviewed studies have shown mean RF in the ranges of -0.27 to +0.20 W m⁻².

Keywords: Biogenic secondary organic aerosols; Day and nighttime atmospheric chemistry; Volatile organic compounds; Cloud condensation nuclei; Tracers.

1. Introduction

Aerosols play important roles in atmospheric process via scattering and absorbing solar radiation, influencing cloud formation and participating in heterogeneous chemical reaction, thereby affecting the distribution and abundance of atmospheric trace gases (Andreae and Crutzen, 1997; Haywood and Boucher, 2000; Choi et al., 2015). Moreover, they affect the radiative balance in the Earth's atmosphere and ultimately the climate (Ramanathan et al., 2001; Novakov and Penner, 1993; Ram et al., 2012; Pani et al., 2016; Tsay et al., 2016; Malavelle et al., 2019; Mahilang et al., 2020). Atmospheric aerosols are categorized into two parts based on their sources; anthropogenic (derived from combustion activities using biomass, fossil fuels, and household solid fuels) and natural (volcanic eruptions, wind-driven soil re-suspension, and mineral dusts (Majumdar et al., 2012). The secondary aerosol particle is reported to be formed by the gas-to-particle conversion processes, nucleation, condensation, and various multiphase chemical reactions (Moffet and Prather, 2009; Riemer and West, 2013; Deshmukh et al., 2016 Mahilang and Deb, 2020).

Plant produced biogenic volatile organic compounds (BVOCs) are extremely reactive in the troposphere with their lifetime ranging from minute to hours. When BVOCs come in contact with atmospheric oxidants the formation of SOA occurs via stepwise heterogeneous reactions (Poschl, 2005; Kawamura et al., 2017; Gunsch et al., 2018; Sinha et al., 2019; Tajuelo et al., 2019). Plants emitting significant monoterpene and isoprene are reported to belong to the families of flowering plants, ferns, conifers, and bryophyte plants (Harley et al., 1999). Some plant species, known for higher emissions of VOCs (emission rate $>5 \mu\text{g g}^{-1} \text{ dry leaf h}^{-1}$), are *Ficus religiosa*, *Adhatoda vasica*, *Eucalyptus species*, *Adhatoda vasica*, *Casuarina equisetifolia*, *Syzygium jambolanum*, *Psidium guajava*, *Lantana camara* (Padhy and Varshney, 2005). Out of

the total BVOCs emissions, isoprene comprises nearly 50% of it; other emitted BVOCs are monoterpenes and sesquiterpenes. The BVOCs undergo photochemical degradation to form gas phase oxidation products and as a result of various heterogeneous reactions of gaseous precursors, SOA are formed, i.e., called biogenic secondary organic aerosols (BSOA) (Xu et al., 2015; Marais et al., 2016; Boy et al., 2019). The extent of BVOCs emission in the atmosphere generally depends upon the geographical location and meteorological conditions (Kourtchev et al., 2008; Helmig et al., 2013). The geographical sites which have sunny, warm tropical conditions and high vegetation biomass density (including plants such as oak, coniferous plants, pine trees), release greater BVOCs in the atmosphere. Above are the reasons why most of the studies were concerned with atmospheric chemistry in sub-tropical areas and the forests (Rissanen et al., 2006; Hoyle et al., 2011). Goldstein and Galbally (2007) reported the measurement of around 10,000-1,000,000 different organic compounds in atmosphere. The VOCs undergo several stepwise degradation processes and further lead to formation of extremely low volatile organic compounds (ELVOC). Many researchers have reported building of, an intermediate, highly oxidized multifunctional (HOM) compound in the gaseous phase of troposphere during the SOA formation process (Kulmala et al., 1998; Ehn et al., 2012). ELVOC compounds are of extremely low volatile nature due to the presence of very large number of functional groups (O/C ratio > 0.7) in HOM causing very low volatility (Mutzel et al., 2015). Jokinen et al. (2014) reported formation of gas-phase HOM, an important channel of monoterpene oxidation, via autoxidation of alkyl peroxy radical by intramolecular hydrogen atom shift. It is believed that HOM may contain many hydroperoxide groups (Mutzel et al., 2015).

Once SOA is formed in the particle phase its fate in the atmosphere continues with the particle growth, aging and ultimately removal either via volatilization or dry and wet deposition processes (Wong et al., 2019). SOAs form HOM organic compounds as intermediates throughout its life cycle (Ehn et al., 2014, 2012; Mentel et al., 2015; Mutzel et al., 2015). At the beginning, BVOC forms the primary products as ELVOC that is detectable using online measurement and characterizable using mass spectrometer. The ELVOC are then condensed into pre-existed atmospheric particles, onto which the rest of the process of SOA formation occurs. Isoprene derived SOA formation in the atmosphere was not considered until 2-methyltetrols was first found in atmospheric aerosols of Amazon forest in 2004. Thereafter many chamber experiments were performed worldwide to determine the SOA composition (Edney et al., 2005; Ion et al., 2005; Clements et al., 2007; Song et al., 2019). Pinic and pinonic acids (α and β pinene SOA tracers) were also detected in the atmospheric aerosol samples worldwide (Yu et al., 1999; Sheesley et al., 2004; Bhat et al., 2007).

The review initiates with a current state of knowledge of the atmospheric reaction mechanism of BVOCs released and the altering overall SOA budget. The next part of the review throws a light upon the oxidation mechanism for SOA precursors and the analytical techniques used for characterization of biogenic aerosols. The review also discussed condensed phase reactions, sulphate aerosols interactions, cloud condensation nuclei (CCN) and future recommendations. There are lots of chamber studies and field works done regarding BSOA but understanding the dynamics of formation and characterization of BSOA are still challenging and also it is important to comprehend their indirect effects in climate and human health (Oberdorster et al., 2005; Mauree et al., 2019). Thus, the objective of this review is to discuss about the recent advancements based on earlier works of BSOA.

2. Atmospheric chemistry of secondary organic aerosols

2.1. Formation of SOA via degradation of VOC

The degradation of the emitted VOC is initiated either by the oxidants present in the atmosphere viz. OH, NO₃, O₃ radicals or by the photolysis. Under certain condition chlorine (Cl) radical also initiates oxidation of VOC. In geographical locations with low amounts of Cl radicals in the atmosphere, the VOC oxidation proceeds via OH initiated mechanism. The Cl radical is believed to be evolved from heterogeneous reaction over sea surface. Many researchers have reported high Cl loading over continental areas (Wennberg et al., 2018, Guo et al., 2020). Hence, the Cl radical initiated VOC degradation completely depends on the geographical locations (availability of Cl radical in the atmosphere). These competitive oxidation processes are either dependent on the nature of parent VOC or on the atmospheric condition (Atkinson and Arey, 2003; Bianchi et al., 2019). Stepwise degradation has been predicted in which the initial oxidation promotes the formation of first-generation products, which includes a set of organic products containing single or multiple oxygenated functional groups like, -CHO, -OH, >CO, -NO₂, -COOONO₂, -COOH, -OOH and -COOOH, etc. A generalized mechanism of SOA formation from VOCs is shown in Fig. 1. The further oxidation of these compounds results in the production of high volatile and soluble second-generation organic compounds, in which further oxidized functional group are added. The lower chain oxygenated products are more volatile than higher or long chain products. The fate of such organic compounds is decided by three stepwise processes viz., fragmentation to CO₂, functionalization to form lower volatility products or by deposition either wet or dry (Atkinson et al., 2006). Many researchers have found the traces of organosulfates and oligomers as SOA in lab and field research both.

Different types of oligomers can be formed over aerosols, such as association of low carbon containing species (oxidation products of glyoxal), epoxides of isoprene and high carbon containing precursors (pinene oxidation products) (Hall and Johnston, 2011). Lin et al. (2014) also reported oligomers formation in aqueous phase glycolaldehyde, glyoxal and methylglyoxal via oxidation of these gaseous species with NO_x and OH.

Organosulfates are sulfate derivatives of SOA and its formation increases as the acidic content of aerosols increases. Surratt et al. (2007) reported that increasing acidity of sulfate seed can significantly enhance the sulfate derived SOA formation. They reported organosulfates formation from C₅-alkene triols, 2-methyltetrols, 2-methylglyceric acid (2-MGA), hydroxyacetone, glyoxal, methylglyoxal and methacrolein present in aerosols. The volatility of the formed product is not only the responsible factor but also some atmospheric parameters like, water contents, cations, anions and relative humidity play vital role in the chemistry of SOA formation (Dommen et al., 2006; Surratt et al., 2006; Denkenberger et al., 2007). There are myriad of organic compounds in the atmosphere which grow or degrade. The most studied aspect of degradation chemistry of the VOC is the OH[•] (OH radical) initiated chemistry which is dominant in the day time (Pagonis et al., 2019). The OH[•] initiated mechanism is mostly preferred for the SOA formation in which radical forms of RO₂ (Peroxy) and RO (Oxy) are reacted wherein RO is formed from RO₂ in presence of high NO_x in the atmosphere. It is also concluded that the formation of product is strongly dependent upon the structure of emitted VOC and also meteorological variables, i.e., temperature and relative humidity etc. (Atkinson et al., 2007). There are three routes by which RO reacts, (1) formation of carbonyl groups and HO₂, (2) decomposition via formation of smaller carbonyls and radical of organic compounds, (3) formation of hydroxy derivatives of carbonyls and HO₂ via isomerization followed by hydrogen

atom shift reaction. NO forms RONO₂ on interaction with RO₂, whereas NO₂ on reaction with RO₂ leads to the formation of atmospheric stable peroxy nitrates (RO₂NO₂). NO₃ plays crucial role in nighttime atmospheric chemistry, whereas O₃ acts as an oxidant both in day and night hours. Somewhere, especially in marine atmosphere, Cl[•] acts as active oxidant and participates in VOC degradation (Guo et al., 2020).

2.2. Effect of anthropogenic pollutants on VOC degradation

Various studies show that atmospheric chemical reactions of degradation mostly depend on the emission strength of VOC in particular geographical location, temperature, and availability of oxidants (Zhou et al., 2019). Increasing atmospheric aerosols can also trigger the condensation of gaseous species onto particle-phase, e.g., BVOC derived gas-phase products are easily condensed over particles phase. Hence, anthropogenic pollutants can significantly enhance the extent of BSOA formation. Another phenomenon that explains the enhancement of BSOA is new particle formation occurring from sulfuric acid and gaseous organic pollutants. Condensation of organic gaseous species over new particles also indicates the growth mechanism of atmospheric aerosols via anthropogenic emission (Bonn et al., 2008; Boy et al., 2008). Hence, the formation of new particles also provides a path for enhanced BSOA formation. The extent of sulfur-containing species can also alter the SOA formation. Xu et al. (2015) reported that isoprene derived SOA (SOAI) is mediated by the atmospheric sulfate loading rather than aqueous contents and aerosols acidity. The NO_x mostly contributes nighttime atmospheric chemistry and trigger the formation of nitrate-containing SOA species of monoterpenes. They reported that NO_x and sulfate alone mediated 43–70% of total OA at the southeastern US during the summer season and suggested that the reduction in emissions of sulfate and NO_x may help reduce the SOA burden. The anthropogenic pollutants affect the transformation of biogenic VOC and

formation of SOA through several mechanisms at low NO_x condition. The vapor phase atmospheric reaction of the pollutants, viz., SO₂ with peroxy radicals promotes the formation of sulfate aerosols, which changes the acidity and water contents and hence coupling occurs between the product formed from biogenic VOC and sulfate aerosols. This coupling results in the regional cooling in the southwestern United States (Zhu et al., 2019). The pathways of reaction as well as the viscosity are affected via the meteorological conditions and presence of oxidants in the atmosphere. As a result, the particle surface available for condensation increase and hence the phenomenon promotes myriad of atmospheric reactions.

2.3. Effect of biomass burning on SOA

Huge amounts of atmospheric pollutants are released globally from biomass burning that contributes around half of the overall worldwide emissions (Bond et al., 2013). The optical properties and cloud processing efficiencies of these biomass burning emissions significantly influence the radiation budget of the Earth and affect the global climate (Ramanathan et al., 2001; Vakkari et al., 2018;). Smoke plumes emitted from biomass burning emissions, which significantly harm the atmosphere, contain carbonaceous components, water-soluble ions, carbonyls, and sugar alcohols (Pani et al., 2019, 2018; Thepnuan et al., 2019). In northern Southeast Asia, significantly high concentration of carboxylates, carbonyl and aliphatic carbon derivatives was found in the smoke released from biomass burning (Popovicheva et al., 2016). A lab study found methylfuran that was produced from epoxides of isoprene under low NO_x conditions (Allan et al., 2014). Methylfuran was also found in field research study, conducted by Hewitt et al. (2010) during the Southeast Asian tropical rain forest campaign. Kelly et al. (2018) reported global annual SOA contribution of isoprene, monoterpene, biomass combustion, and anthropogenic emission to be 19.6, 19.9, 9.5 and 24.6 Tg (SOA) a⁻¹, respectively, during 2017.

Enhanced global SOA production was reported due to the photochemical degradation of biogenic and biomass-derived precursors. The aging processes and SOA formation from non-methane organic gases are greatly influenced by OH exposure that regulates the heterogeneous oxidation reaction during SOA formation (Pandey et al. 2018; Lim et al., 2019; Pandey et al., 2020). Significant atmospheric warming and surface cooling was reported due to enhanced biomass burning emissions (organic carbon ratio > 90%) with high loading of NO_3^- , OC3 and EC1-OP; ultimately leading to a severe haze in northern peninsular Southeast Asia (Pani et al., 2018). Thepnuan et al. (2019) obtained a good correlation between NH_4^+ and SO_4^{2-} , as an indication of secondary formation, and established a link between biomass burning and high correlation between NO_3 and levoglucosan in the upper South East Asia region. They also found high loading of oxalate ions due to biomass combustion. Vakkari et al. (2018) found a significant enhancement in SOA formation and doubled PM_{10} loading due to biomass burning emission in southern African fires. Ciccioli et al. (2014) reported about the release of β -caryophyllene, which resides over wood and leaf surface due to low volatility, upon the burning of biomass. A strong correlation between β -caryophyllene and levoglucosan also indicated an association between biomass burning and secondary formation (Wang et al., 2019).

2.4. Day time atmospheric chemistry

2.4.1. OH^\cdot Initiated mechanism

During daytime high temperature with intense sunlight enhances the biogenic VOC (isoprene and monoterpenes) productions, i.e., high SOA loading are generally found during daytime. Li et al. (2019) found that average atmospheric temperature is strongly correlated with isoprene ($R^2 = 0.84$) and monoterpenes ($R^2 = 0.91$) emissions in the atmosphere. A study conducted at coniferous forest found that the concentration of hydroxyl radical was

comparatively higher (10^5 - 10^6 molecules cm^{-3}) during daytime. Hence, OH radical initiated chemistry dominates during daytime (Bates et al., 2019). When OH radicals are formed and interact with biogenic VOC (emitted from the vegetations), a series of oxidation reaction get started, and the biogenic VOC are degraded to produce low volatile organic compounds (products). Fig. 2 displays atmospheric degradation pathway of biogenic volatile organic compounds. Fig. 3 shows the detailed mechanism of gas phase oxidation of isoprene as an important constituent of VOC. The oxidation of isoprene by OH radical at low NO_x concentration produces hydroxyhydroperoxide that further produces isoprene epoxydiol (IEPOX) as an important intermediate in atmosphere reactions (Xu et al., 2015). Then further degraded RO_2 radical is produced, which further yields carbonyl and hydroxycarbonyls. The fate of these RO_2 radical is dependent on NO_x (mixture of NO and NO_2) concentration. High BSOA with low NO conditions yield RO_2 radical that further undergoes transformation to RO_2NO_2 and RONO_2 . This finally ends up to alkoxy radicals and smaller volatile fragments. However, at elevated NO_x conditions methanecaryloperoxynitrates (MPAN) are formed as intermediate, which tend to generate a mixture of product dominated by peroxy acetyl nitrate (PAN), carbonyls, hydroxyl carbonyl and organic nitrates (Liu et al., 2019a). PAN, an important constituent of the photochemical smog, is organic peroxy nitrate that is formed by the reaction of RO_2 radical and NO_2 .

2.4.2. O_3 initiated mechanism

Ozone gas generally comes to the troposphere from stratosphere via eddy flux processes. Besides this, it can also be formed via photochemical reaction between VOC (biogenic and anthropogenic) and NO_x (Calfapietra et al., 2013). Due to high isoprene emission flux and formation of ozone during the daytime, it can be stated that the ozone degradation dominates at

day time. Ozone is also produced by photochemical oxidation of CO, CH₄ and non-methane VOC in presence of NO_x. The loss of tropospheric O₃ takes place through dry deposition and various other chemical reactions. The term photochemical indication, used in order to evaluate the extent of ozone formation, depends upon the concentration of H₂O₂, HNO₃ and reactive nitrogen species (Gen et al., 2019). The chemistry initiated by O₃ tends to feed into similar mechanism as those of NO₃ and OH[•] by virtue of the formation of OH, RO₂ and RO radicals (Faiola et al., 2019).

Johnson and Manstson (2008) described that O₃ initiated chemistry is generally believed to take place by Criegee intermediates (carbonyl oxides) formation. These intermediates are formed by decomposition of primary ozonides of isoprene via two pathways to give a carbonyl along with it. A carbonyl substituted Criegee intermediate can be also formed through α -pinene (Zhou et al., 2019). Initially formed energy rich Criegee intermediate gives further reaction by a number of pathways, i.e., A) collisional stabilization of thermalized Criegee intermediate and, B) for dialkyl and monoalkyl substituted Criegee intermediate, isomerisation to a 'hot' hydroperoxide followed by decomposition (Atkinson and Arey, 2003). In the stepwise photooxidation of isoprene and α -pinene, generally, epoxides are observed as end product (Ng et al., 2006). Ozone alkene reaction system involves both OH radical and O₃ as oxidants (Atkinson et al., 1993). At the end of the process, product distribution includes carboxylic acid and hydroxyalkyl hydroxyperoxide, which are sensitive to both NO_x and H₂O and also to scavengers such as carbonyl and acid scavengers (Hallquist et al., 2009).

2.5. Nighttime atmospheric chemistry

2.5.1. OH[•], and NO_x initiated mechanism

During night time, the levels of the OH radical is almost minimized, hence the nighttime chemistry is mainly dominated by NO_3 radicals, which are formed via reaction of NO_x with ozone. Further, NO_3 reacts with NO_2 to form N_2O_5 and establishes equilibrium between them, thus both NO_3 as well as N_2O_5 affect degradation at night. Ng et al. (2008) performed chamber experiments and observed that the organic nitrates were formed from reaction between NO_3 radical and BVOC. However, along with NO_2 and NO the OH radicals are also formed even at night upon decomposition of nitrate substituted peroxy radicals, which also contribute in degradation of VOC. The oxidation products (SOA) of NO_3 are mainly multifunctional organic nitrates containing carbonyl, alcohol and peroxide groups or of compounds of lower vapor pressure. The N_2O_5 initiated chemistry also operates and much of its chemical reactions completes on the particle phase of aerosols.

The formation of ammonium nitrate in the ambient atmosphere accounts for the removal of NO_x from the atmosphere. This nighttime mechanism can remove as much as NO_x as the day time gas phase reaction with OH[•] with NO_2 to produce HNO_3 . Nighttime loss of N_2O_5 thus limits the availability of NO_2 for the photochemical production of O_3 the following day (Xia et al., 2019).

At high NO_x condition RO radicals are formed, the fate of which is dependent on the structure of parent VOC. All these degradations can result into carbonyl, HO_2 or it may also decompose through C-C bond breaking via formation of smaller carbonyls and organic radicals. Isomerization through H-atom shift may also take place to produce hydroxy carbonyl compounds and HO_2 . When OH radical interacts with isoprene then there are possibilities of both self and cross reactions to take place. The later results 1,2-diols formation in the natural tropical

atmosphere at both low as well as high NO_x conditions during high biomass burning activity (Anthropogenic activity) (Liu et al., 2019a).

Surrat et al. (2006) investigated the chemical composition of products formed by isoprene oxidation both under high and low NO_x condition and concluded that formation of high MW product was dependent on the concentration of NO_x. At low NO_x condition organic peroxides were produced and significantly contributed to the SOA mass, which further decreased with time and thus also indicated photochemical aging. The major products were 2-methyltetrols, C₅-alkene triols, and the minor products identified were 2-methyltetrol oxyformate derivatives, hemiacetal derivatives and C₅-trihydroxyl monocarboxylates. These compounds were also identified in the PM_{2.5} (matrix) in ambient air matrix of Amazonian forests. Different BSOA tracers with their precursors and mass fragments are given in Table 1. At high NO_x condition, major products formed are acidic oligoesters with 2-methylglyceric acid as a key monomeric acid unit and residues formed by esterification of OH groups with acetic/ formic acid (Cui et al., 2019; Surratt et al., 2006). Table 2 illustrates different studies regarding BSOA and their concentration levels in the ambient atmosphere.

2.6. Role of ammonia in SOA formation

The extent of ammonia (NH₃), generally present as a trace gas in troposphere (Zaveri et al., 2020), depends upon the source and strength of emission in the atmosphere (Li et al., 2020). NH₃ in the atmosphere comes from nature as well as anthropogenic sources. NH₃ can be emitted mostly from livestock like cows, beef, chicken and hogs (Yu et al., 2020; Zhang et al., 2020a). However, NH₃ from mobile emission sources and soil emissions are also significant (Zhu et al., 2020). The atmospheric nitric (HNO₃) and sulfuric acids (H₂SO₄) react with NH₃ and form

NH_4HSO_4 , NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$ (Wang et al., 2020a). The fate of atmospheric NH_3 is generally finished by OH radical initiated oxidation, wet and dry deposition (Aguilar-Dodier et al., 2020). NH_3 is less reactive in the troposphere, hence its removal via gas phase oxidation process is less significant (Fairlie et al., 2020). Its reaction with HNO_3 and H_2SO_4 leads to formation of secondary inorganic aerosols (Agarwal et al., 2020). Researchers also reported the interaction of atmospheric NH_3 with organic acids leading to enhanced SOA formation (Du et al., 2020). SOA formation from NH_3 occurs via formation of condensable salt formation through acid-base atmospheric reactions (Jang et al., 2020). Organic aerosols are ubiquitous in nature and covers major fraction of atmospheric aerosols (Wu et al., 2020). An earlier work has reported that fog and hazy atmosphere include significantly high organic aerosols loading due to their highly polar nature (Izhar et al., 2020). Acetic and formic acids are well known conventional organic acids present in gas and particle phase in the atmosphere (Link et al., 2020). These organic acids are derived from biogenic as well as anthropogenic sources. Among the various organic acids, dicarboxylic acid is formed via oxidation reaction between atmospheric hydrocarbons and ozone (Zhang et al., 2020b), and the pinic and pinonic acids are formed from oxidation reactions of α -pinene. Researchers have reported that the pinic acid is the most abundant species in particle as well as gas phase in the atmosphere (Babar et al., 2020; Kołodziejczyk et al., 2020). The organic acids produced in gaseous phase react with NH_3 and form condensable salts (Qi et al., 2020). This phenomenon indicates the enhanced SOA formation in NH_3 rich atmosphere (Sorooshian et al., 2020). Some exceptional cases are also reported in which acetic and formic acids do not form condensable salts with NH_3 (Abbatt and Wang, 2020).

In one of the chamber studies, α -pinene was primarily allowed to react with O_3 until the aerosol formation and thereafter NH_3 was added in the reaction chamber (Bagchi et al., 2020). This caused significant enhancement in the numbers and aerosol mass concentration and indicated the triggering of gas-to-particles conversion process in the reaction chamber. They observed that adding NH_3 enhanced the particles size from 242 to 248 nm, which indicated the condensation of SOA (formed from α -pinene, ozone and NH_3) over pre-existing particles. Similarly, enhanced SOA (e.g., pyruvic acid and 2-methylglyceric acid) formation was reported during the reaction of VOC (isoprene) with ozone and NH_3 (Chen et al., 2020; Wang et al., 2020b). Firstly, energy rich ozonide is formed via oxidation reaction of double bond of alkene with ozone. The ozonide is then decomposed in carbonyl substituted C10-C1. The norpinic acid, pinonic acid, pinic acid and norpinonic acid were found to be produced via α -pinene, ozone and NH_3 reactions (Winterhalter et al., 2003; Lengyel et al., 2020).

Relative humidity also plays crucial role in SOA formation (Sun et al., 2013; Cheng et al., 2015). Fick et al. (2003) reported that formation of pinonaldehyde and pinic acids are significantly enhanced at higher relative humidity. However, pinonic acid formation decreases with higher relative humidity (Kammer et al., 2020). They also reported that the SOA yield under dry condition is higher than the yield obtained for humid condition. However, a kinetic modeling study conducted by Kamens et al. (1999) reported that water vapor increased the gaseous SOA formation and condensation over particle phase. The enhanced SOA formation under humid conditions are significant than dry conditions (Liu et al., 2020; Pratap et al., 2020). Humid conditions are also reported for acidic SOA product formation (Geng et al., 2020). To investigate the role of acidity, the oxidation reaction of VOC with ozone and NH_3 was performed at pH 9.0 (Jang et al., 2002). High SOA yields were obtained under humid (31%) and acidic

conditions. The proposed reasons were increasing in gaseous organic acid and subsequent condensation and particle phase SOA formation under NH_3 rich environment.

2.7. Effect of acidic aerosols

Atmospheric aerosols may have acidic nature depending upon the concentration of water-soluble inorganic ions. These acidic aerosols have significant impact on atmosphere like lower visibility, cloud processing and human health impacts (Zhang et al., 2007). Increased SOA loading is also reported in many works due to the presence of acid catalyzed aerosols (Tsigaridis and Kanakidou, 2003). A chamber study of reactions, between isoprene and oxidants at different $[\text{H}^+]$ acidity, was conducted by Surratt et al. (2007) towards finding the influence of aerosol acidity on isoprene derived SOA formation. They found that the obtained mass of the SOA was linearly correlated with acidity, hence concluded that higher acidity caused more SOA formation. Yields of 2-methyl erythritol and 2-methyl threitol present in particle phase were also found to increase with increasing acidity. High isoprene derived SOA with acidic aerosols is also reported in ambient air (Jaoui et al., 2004). Formation of one of the isoprene products, i.e., 2-MGA, however, was reported to be unaffected by acidic aerosols (Edney et al., 2005; Kleindienst et al., 2009). In another study, Zhang et al. (2007) reported a 25% increase in secondary organic carbon yield with high acidity of ambient aerosols at Pittsburgh. They reported the acid catalyzed effect on SOA formation. Offenberg et al. (2009) conducted a laboratory study of SOA formation derived from β -caryophyllene and α -pinene by varying $[\text{H}^+]$ concentration and keeping NO_x and precursor concentration constant at simulated condition of ambient atmosphere. Isoprene and β -caryophyllene both were allowed to oxidize with NO_x onto smog chamber. The SOA yields were reported to be high with increasing aerosols acidity for β -caryophyllene, while inverse

results were reported for α -pinene derived SOA yield which showed less SOA yield with high aerosols acidity.

2.8. Gaseous and liquid phase interactions during SOA formation

Atmospheric gases and their uptake towards liquid phase play key role in the extent and nature of SOA formation (Gorkowski et al., 2020). Most of the reactions occur at the interface between the atmospheric gases and liquid phase of aerosols (Qian et al., 2019). Brüggemann et al. (2020) reported about inadequacy of the only liquid phase mechanism to explain SO_2 initiated oxidation mechanism, as it occurred via bound complex formation between gas and liquid interface. Zhang et al. (2019a) also reported that glyoxal and acetaldehyde uptake mechanism also undergo complex reactions at the interface. Enhanced reactivity at the interface is reported due to photochemical activity. A study reported that presence of light between 300-500 nm strongly enhances the NO_2 initiated SOA formation process due to photosensitized transfer of electron (George et al., 2015). The photo induced reaction is stronger on humic acid in which a light induced reduction of NO_2 leads to gaseous nitrous acid (HONO) formation. The chambers experiment also indicated the HONO formation on humic acid aerosol. A similar chambers experiment revealed the occurrence of a photo induced reaction with strong absorption band appearing at 450 nm for benzophenone and phenol when exposed to ozone (Zhang et al., 2020b). The similar shift of band was also observed for photochemical oxidation of limonene during the SOA formation process in the atmosphere (Shiraiwa et al., 2019). Chambers experiments revealed that carbonyls undergo aldol condensation in sulfuric acid containing solution forming oligomers (Mekic et al., 2019). The ammonium sulfate and amino acid in aqueous phase were found to form light-absorbing “humic-like” substances (Grace et al., 2019).

2.9. Ice nucleation

Cloud phase reactions and ice nucleation both play crucial roles in maintaining the atmospheric radiation budget of the Earth (Quante, 2004). Earlier works have reported two types of nucleation phenomena, i.e., homogeneous and heterogeneous nucleation during ice nucleation (Spichtinger and Gierens, 2009). Inorganic salts, dust particles, and bioaerosols are believed to trigger the ice nucleation activity (Baustian et al., 2013). Some organic aerosols like oxalic acid, citric acid, and ethylene glycol have been reported as ice-nucleating species (when present as single-component organic aerosol) (Murray et al., 2010). Organic aerosols can exist as liquid, amorphous and glassy forms depending on the composition and water extents of aerosols (Koop et al., 2011). A laboratory study conducted by Mohler et al. (2008) found that α -pinene derived SOA is effective towards ice nucleation. Another work, however, has reported that the α -pinene SOA shows weak ice nucleation activity in the cirrus cloud presents at an upper troposphere (Ladino et al., 2014). Wilson et al. (2012) and Schill et al. (2014) found SOA and organic aerosols to be sufficiently active towards ice nucleation. The mixtures of dicarboxylic acid, raffinose, and ammonium sulfate seed aerosols are reported to be highly active towards ice nucleation at temperature <220 K (Wagner et al. 2012). SOA containing methylamine and methylglyoxal presents as glassy state and shows poor ice nucleation activity. However, adding ammonium sulfate significantly enhances their ice nucleation activity. Ammonium chloride and ammonium sulfate coated with long-chain alcohol have been found to increase the freezing temperature of solution (Cantrell and Robinson, 2006; Zobrist et al., 2008). Like the structure of ice, the alcohol-containing aerosols can assemble into 2dimensional crystal at the air-solution interface. Prenni et al. (2009) have demonstrated the none-ice nucleation-activity of 25 different alkenes molecules containing SOA at 243 K. Apart from these works, there is still a substantial

knowledge gap between the SOA and ice nucleation activity. Hence much research is needed to improve the understanding of SOA formation and ice nucleation.

3. Analytical techniques employed

3.1. Molecular characterization of biogenic constituents of SOA

When molecular characterization of particular compound is done, a term tracer or marker is used that allows the chemists to gain insight into the composition of complex matrix such as the ambient aerosols (Johnston et al., 2019). In the photooxidation of isoprene, 2-methyltetrols, 2-methylglyceric acid, C₅-alkene triols and related oligomer products have been identified as tracers and have been also confirmed in the field samples from the Amazon basin, Brazil and from mixed coniferous and deciduous forest in Hungary (Claeys et al., 2004). The GC/MS with prior trimethyl silylation was used for analysis of ambient aerosol samples. The molecular formula of constituent SOA tracers can be estimated using chemical ionization mass spectrometry (CI-MS) (Wang et al., 2004).

3.2. Analytical advancements

The SOA consist of a myriad of compounds, which exhibit diversity in their physical nature in atmosphere and hence the techniques involved in their complete characterization become complicated. Based on the capability of acquiring information on the characteristics of SOAs, the techniques employed may be explained into following categories- indirect method, online technique, and the offline technique. The difference between measured total organic aerosols and estimated primary organic aerosols (POA) forms the basis of indirect method of quantification of SOA. The offline measurement technique includes nuclear magnetic resonance spectrometer (NMR), gas chromatography-mass spectrometry (GC-MS), Fourier transform infra-red

spectrometer (FTIR) and liquid chromatography-mass spectrometry (LC-MS), which give complete information about different chemical components of organic aerosols. Aerosol mass spectrometer (AMS) comes under online measurement technique that provides fast acquisition time and near real time data but they are less capable of providing information about composition (without providing details of individual fractions) (Ide et al., 2019).

The most widely used technique for characterization of SOA from complex mixture aerosols is gas chromatography coupled with mass spectrometry, which is utilized as the most powerful separation and analytical technique (Kotianova et al., 2004). The hyphenated techniques like GC-MS is usually used with electron ionization (EI) or chemical ionization (CI) detectors and similarly, LC-MS is used with atmospheric pressure chemical ionization (APCI) and electrospray ionization (ESI) detectors in positive and negative ion modes. The complexity and highly oxidizing nature of SOA make some limitation to completely characterize individual components, because they can give overlapping peaks and may cause majority of mass fractions as unresolved (Williams et al., 2007). To overcome such situations, researchers have developed derivatization process that makes organic compounds available for chemical characterization using analytical techniques (Claeys et al., 2004, Jaoui et al., 2005, Szmigielski et al., 2007, Healy et al., 2008). The molecular mobility in the GC column is generally restricted by the presence of alcohol, carbonyl and carboxylic acid groups in the molecules (Liao et al., 2007). Poor detector response is obtained in GC due to interaction of such molecules with stationary phase. Moreover, some chemical species remain undetected. Derivatization process includes formation of derivatives of analytes. Derivatives formation suppresses polarity and increases volatility of the analytes. Hence, derivatization improves separation of mixtures and provides better GC response. Derivatization for GC analysis is done via three different reactions, i.e., acylation,

alkylation and silylation (Flores et al., 2014). Simply in the silylation reaction, insertion of a substituted silyl group occurs. The hydrogen atom replacement via silyl group increases polarity of less volatile molecules and, hence, it makes the analytes suitable for GC separation (Flores et al., 2015). To increase the resolution, researchers have developed two dimensional GC coupled with time of flight (TOF) mass spectrometer that has the capability of detecting 10,000 organic species in the ambient atmosphere (Hamilton et al., 2004).

For characterization of high molecular weight and highly polar compounds LC is used. Mainly, LC is used for analysis of carboxylic acids (Walhout et al., 2019a). Further, the hyphenated technique, i.e., LC-MS has better detection limit, high sensitivity and good reliability over LC with fluorescence detector technique. LC does not need any derivatization techniques for analysis (Iinuma et al., 2007). To overcome the problem regarding low resolution, Iinuma et al. (2007) used two dimensional LC for characterization of organosulfates in pinene derived SOA.

Fourier transform infrared (FTIR) spectroscopy is used for the detection of functional groups in compounds, in samples collected on filters. One of the major advantages of this technique is that it is capable of measuring some functional groups, e.g., amines and organosulphates, which are difficult to measure using other techniques. Many researchers have employed FTIR spectroscopy in field samples (Takahama et al., 2019; Wallington et al., 2019). The OA characterized by FTIR spectroscopy is also in good agreement with AMS study in many reported works (Lechevallier et al., 2019; Singh et al., 2019).

Very high-resolution techniques such as ultra-high-performance liquid chromatography electron spray ionization quadrupole-TOF-MS (Particle into liquid sampler (PILS) - Ultra

performance liquid chromatography (UPLS) / ESI-Q-TOF-MS) in liquid sampling has been used for quantification of SOA. TOF/MS is used for differentiating secondary volatile organic compounds (SVOC) and particle phase species. The SOA are firstly derivatized by silylation and thereafter separated by GC enabling sequential identification and quantitative analysis of alkanolic acid, polyols, diacids, sugars and multifunctional compounds with MS detector. A good time resolution online technique for quantification of SOA includes AMS for quantitative measurements of time resolved aerosol chemical composition of atmospheric aerosols (Salcedo et al., 2006).

A direct determination of glyoxal by open path differential optical absorption spectroscopy (DOAS) has been done by researchers, which presents novel means to get information about VOC oxidation process (Volkamer et al., 2006). Chemical ionization mass spectrometers (CIMS) now-a-days is employed frequently for quantification and characterization of VOC through ionization with reactive ions such as H_3O^+ , NO_3^- , CF_3O^- , or acetate and also for characterizing organic trace gases. The multi orifice volatilization impactor (MOVI) coupled to TOF/CIMS has also been used in some works that provides the molecular mass data, but not fragmentation, to identify isomers or additionally unknown compounds (Nozière et al., 2015). Thermal desorption aerosol gas chromatography (TAG) is also a frequently used technique for separation of the organic molecules from the matrix. Since CI is a soft-technique, instead fragmentation pattern, it only provides information about the molecular mass, hence it is coupled to MOVI flux measurement device to gather more information (Isaacman et al., 2014). Measurement of ambient aerosols with good time resolution is done using aerosol mass spectrometer (AMS). The complicated mass spectra obtained with the use of electron ionization

are resolved into individual oxygenated and deoxygenated organic aerosol components using positive matrix factorization (PMF) source apportionment (Zhang et al., 2011).

The mostly employed analytical techniques used for OA, compared with each other on the basis of the three features: time and size resolution, percent of mass that can be analyzed, and chemical resolution of an instrument, are summarized logically through chess-board scheme in Fig. 4. Here, pre-eminent pieces of chess have been used as a depiction of the most powerful analytical technique. This most powerful piece was considered on the basis of number, and direction of moves and hence the overall ability to attack on the opponent. Here, Queen is considered as the most powerful piece and Pawn is considered as least powerful piece. Here Queen is representing 2D-GC/MS and Pawn is representing the assumed useless analytical technique. The positions of particular piece/analytical technique have their own importance. It can be understood as a 3Dimensional graph, containing three different scales, i.e., mass percent analysed (Y1 scale), chemical resolution (X1 Scale) and time and size resolution (Y2 scale). Hence, the assumed perfect instrument is placed in h8 position while, the useless instrument is kept in a1 position of the chess-board. In the similar way all the analytical techniques are compared using chess-board as 3D representation.

As mentioned in the figure, GC/MS, which provides molecular identity, is able to investigate only 40-50% of the complete aerosols mass existing in the atmosphere. The analysis of organic mixtures using GC/MS, mostly takes 1 hour as reported by William et al. (2007) but the drawback is limited size resolution. Similarly, the thermal-optical carbon analyzer can determine the total OC with 1-hour time resolution but with limited size resolution. However, the combination of techniques will completely be able to provide more about the composition of aerosols and their quantification with better time and size resolution in the analysis of organic

aerosols (Goldstein and Galbally, 2007). PILS-WSOC, which includes the use of particles into liquid sampler (PILS) is available with the analysis of water-soluble organic carbon (WSOC). PILS has the ability to achieve the time resolution of minutes but there is also drawback of limited size resolution. This was not possible with water-soluble organic carbon analyzer alone. The other instrument, AMS provides good time and size resolution, it can also give chemical composition, whereas the characterization done by NMR and FTIR is more directly associated with functional groups (Aiken et al., 2008; Decesari et al., 2007). Hence, the 100 percent perfect instrument with all necessary characteristics does not exist, but hyphenation of techniques is useful for characterization of OA.

4. Worldwide and seasonal variation of BSOA

In this section, the seasonal variation and worldwide estimation of three main categories of BSOA tracers derived from isoprene (SOAI), monoterpenes (SOAM) and sesquiterpenes (SOAS) are described. Zhu et al. (2017) estimated BSOA tracers in $PM_{2.5}$ aerosols of Shanghai, China during summer-spring season. They found the SOAI tracers to be the most abundant over SOAM and SOAS. The SOAI formation was found to be temperature dependent; hence, high concentration of SOAI was reported for summer days. The study revealed a weak correlation indicating the non-dependence characteristics between relative humidity and SOAI, and a significant increase in SOAI formation with increase in the acidity of aerosols. One of the SOAI, 2-methylglyceric acid was reported to have different formation process marked by weak correlation between 2-methylglyceric acid with other SOAI. Interestingly, cis-pinonic acid was reported to be the first oxidation product of monoterpenes based on its negative correlation with all other SOA tracers. Yuvan et al. (2018) discussed the seasonal differences and the meteorological factors influencing SOA formation at Pearl River Delta. SOAI loading was found

significant throughout the study period; however, SOAM and SOAS formation were dominated in the summer season. Atmospheric oxidants also played crucial role in SOAI, SOAM and SOAS formation via heterogenous photochemical reactions onto the particles surface.

BSOA are usually found more abundant in summer over winter. Fu et al. (2016) reported Coastal region Mumbai (India) has high abundance of BSOA in winter than summer. This was reported to be due to the predominant southwest summer monsoon, which circulated clean marine air to Mumbai, India. The rise in SOA abundance and its temporal variation during winter indicated large contributions of continental aerosols via long-range transport along with local sources. Contrastingly, Fu et al. (2016) also found SOAI and SOAM to be more abundant than those of SOAI at Mumbai, India. In another work, organic molecular composition of PM₁₀ samples, collected at tropical region of India, viz., Chennai for BSOA showed the high abundance of SOAM and SOAS in late winter, whereas in summer SOAI contribution was more significant. The abundance of β -caryophyllene SOA tracers were found comparable during winter and summer at Chennai (Fu et al., 2010).

Kang et al. (2018) estimated the molecular composition and concentration of BSOA and found the significant transportation of BSOA from continental inland areas to East China Sea. Here, SOAM was found to be the dominated species over SOAI and SOAS, which indicated the formation of photochemically aged SOA through long-range transport. The concentration of SOAI tracers were reported to be higher than other SOA tracers in aerosols samples of urban site in Jinan North China (Liu et al., 2019b). The SOAI tracer was reported to be high in summer and low in the winter. In this work, the increase in SOAI and SOAM concentration was correlated with increase in atmospheric ozone concentration, and decrease in their respective concentrations with increasing NO_x concentration. In another study Hong et al. (2019) reported high SOAI

abundance than other tracers in PM_{2.5} during summer season at Mt. Wuyi, southeastern China. The reason behind more abundant 2-methylglyceric acid in fall-winter season was concluded to be due to atmospherically favored conditions like low temperature and higher NO_x concentration. Here also β -caryophyllene and levoglucosan were found strongly correlated indicating biomass burning emission as a common source. In another work, Ding et al. (2011) reported an estimate for BSOA tracers in fine particles (PM_{2.5}) over rural region Pearl River Delta (PRD) in south China. Here, 2-methyltetrols showed good correlations with atmospheric temperature. 2-Methyltetrols was also strongly correlated with aerosol acidity, which indicated the enhanced SOAI formation with aerosol acidity. The negative correlations found for α -pinene SOA tracers may produce from their transfer in particle to gas phase with the increase of aerosol acidity. Shen et al. (2018) investigated the PM_{2.5} samples from urban sites like, Beijing, Tianjin, Shijiazhuang and Xinglong (background site) over one-year time duration. The background site, Xinglong had seasonal highest SOAI and SOAM concentration over urban sites. SOAI had more abundance in summer, however, the SOAM and SOAS had more abundance in spring. They also concluded that the background site was mostly influenced by biogenic sources. However, urban sites were mostly affected by anthropogenic sources.

Haque et al. (2016) estimated total suspended particulates (TSP) samples from central Alaska, USA for BSOA. Here, SOAI was reported to be more abundant in summer; however, SOAM and SOAS were found least abundant in summer. Contrastingly, SOAM and SOAS were found more abundant in spring whereas, SOAI was least abundant in spring. Hoque et al. (2016) also reported that the β -caryophyllene was strongly correlated with levoglucosan biomass burning tracers indicating that the β -caryophyllene was also significantly contributing towards

biomass burning. The Subarctic Alaska aerosols were enriched with α/β -pinene and β -caryophyllene, biogenic SOA tracers.

Ren et al. (2019) monitored PM_{2.5} aerosol samples of Mt. Wuyi and observed that the abundances of the isoprene and monoterpene SOA tracers to be higher in summer season than other seasons, whereas the β -caryophyllinic acid was found lowest in summer. Concentrations of BSOA tracers showed a positive correlation with temperature and a negative correlation with relative humidity. Moderate correlations between the gaseous pollutants (SO₂ and NO₂) and BSOA tracers observed in the mountain top of southeast China suggests that the gaseous pollutants trigger the acid-catalyzed heterogeneous reaction and SOA formation. In this work also significant correlation was observed between the β -caryophyllene and levoglucosan indicating their genesis from biomass burning. Ren et al. (2019) also concluded that anthropogenic sources in China may enhance BSOA production in the distant mountain top regions.

5. Environmental impact of SOA

The presence of SOA in the atmosphere has impact on the climate and health. Respiratory diseases, reduced visibility, haze, degradation in the air quality leading to a polluted environment and more are the results of presence of organic species in atmospheric aerosols. The regional cooling observed in the southeastern US have been attributed to them. It is expected that NO_x, SO₂, and VOC emission would be decreasing in North America, Europe and vicinity areas, whereas it would be increasing in Asian continent, in future. These changes in the climate, disturbs the global ecosystem distribution and ultimately BVOC emission rates. Consequently, ozone levels would get affected and thus the SOA formation in both regional and global scales.

The vulnerable arctic environment is under special threats since it has experienced warming at a rate two times the rate of the global average. Moreover, surprising depletion in tropospheric ozone concentration at Polar Regions was reported. The SOA so formed influence the ecosystem in many ways playing a key role in tropospheric chemistry including ozone dynamics, CO production and methane oxidation.

SOA has some optical properties; it absorbs and scatters the solar radiation hence alters the radiation budget of Earth. The optical properties of SOA may depend upon the chemical complexity of atmospheric SOA, presence of atmospheric oxidants, VOC loading and aerosols aging (Moise et al., 2015). Optical properties of climate can be estimated by the parameter named radiative forcing SOA (RFSOA). The annual average RFSOA was reported to be -0.27 to $+0.20 \text{ W m}^{-2}$ (Boucher et al., 2013). This finding is also mentioned in the 5th annual assessment report by Inter-Governmental Panel on Climate Change in 2014. Many of the global climate models have been established to estimate the RFSOA of the organic aerosols in which the interaction between aerosols and radiation due to light scattering and absorbing properties of SOA have been described. Scattering of radiation has cooling effect; however, absorbing nature has heating effect on the atmosphere. Recent model studies have revealed organic aerosols to be mainly white particles having very small absorbing nature (Hoyle et al., 2009; Spracklen et al., 2011; Myhre et al., 2013). Myhre et al. (2013) made a comparative study between 16 global model results and concluded RFSOA value for cooling to be -0.4 W m^{-2} , which was higher than the heating estimates. Results indicated the dominance of scattering SOA on directly influencing the climate. In addition, light absorbing SOA also significantly contributed to influencing the climate. Absorbing SOA is a broad part of organic aerosols present in the atmosphere named brown carbon that absorbs in much stronger wavelength than black carbon (Liu et al., 2015).

Shrivastava et al. (2015) reported that atmospheric burden of SOA is far higher than the black carbon because brown carbon strongly absorbs at lower wavelength. Hence, absorptivity coefficient by SOA has greater importance to evaluate atmospheric implication. Absorptivity on particular wavelength depends upon the precursors of SOA, their formation process and geography of location (Zhang et al., 2011).

Less absorbing SOA ($\text{MAC}_{405\text{nm}} < 0.02 \text{ m}^2 \text{ g}^{-1}$) was formed from oxidation of α -pinene via OH radical, whereas SOA from toluene via OH/NO_x as oxidant was more absorbing ($\text{MAC}_{405\text{nm}} \sim 0.4 \text{ m}^2 \text{ g}^{-1}$) nature (Lin et al., 2015). In addition, absorptivity of atmospheric SOA also depends on the presence of ammonia that increases the absorbing coefficient of SOA (Updyke et al., 2012). On the other hand, Lee et al. (2014) reported that photochemical aging could also alter the light absorbing properties of SOA. Solar radiation can decompose certain absorbing species; however, some light absorbing species may form onto particle phase via heterogeneous photochemical reactions (Hinks et al., 2016). Generally, dehydration of aerosols and cloud particles may form high light absorbing oligomers. However, water uptake may result hydrolysis and less light absorbing aerosols (Nguyen et al., 2015). Additionally, photochemical reactions of imidazole on particle phase also alter the optical properties of aerosols by influencing SOA formation and aging process (De Haan et al., 2019).

Furthermore, absorption coefficient of SOA via other sources, like biomass burning is also significant. Biomass burning is considered to be the major source of atmospheric brown carbon (Washenfelter et al., 2015). Zhang et al. (2016) also found the absorptivity of primary biomass combustion aerosols (405nm , $\text{BBOA} \sim 0.5\text{--}1 \text{ m}^2 \text{ g}^{-1}$) to be far greater than the absorptivity of SOA. Recent global modelling studies have focused on the potential importance of primary and secondary OA from different sources (Wang et al., 2016; Lin et al., 2015; Wang et al.,

2014). These studies also incorporated regional measurement of optical depth of aerosols and satellite-based measurement to find global brown carbon contribution. It was concluded that the heating effect was regionally influenced by the absorbing organic aerosols from biomass combustion and non-natural emissions was comparable with evolved SOA cooling. Ultimately the absorbing nature of SOA depends upon the chemical complexity, formation mechanism, their sources and atmospheric aging.

6. Recent advances in atmospheric chemistry

Researchers across the world have focused on the novel SOA formation mechanism, cloud aerosol interaction and their environmental impacts. Garofalo et al. (2019) investigated the aerosols released from forest fire of western United States and concluded that those aerosols were the products of subsequent oxidation and condensation over particles. Faiola et al. (2019) studied the impact of stress caused by insect's attack on pine trees and their association with VOCs emissions. They also reported significant changes in VOC emissions from plant due to this stress caused by insects. Recently, laboratory research works have revealed the interaction of biomass derived 3-methylfuran and resorcinol with NO_3 radical and subsequent SOA formation in aerosols (Finewax et al., 2019). Similarly, organic nitrates containing SOA formation was reported from multistep oxidation of Δ -3-carene (Draper et al., 2019). In another work, particle phase hydrolysis reactions have been studied in the atmospheric aerosols by Zare et al. (2019). It was concluded that organic nitrates SOA are no longer identifiable due to its hydrolysis onto particles phase. Walhout et al. (2019b) studied about the photochemical aging process of pinene derived SOA. They used the combination of FTIR and UV-vis studies with the offline mass spectrometry to investigate the SOA formation. They also reported that the residence time of aerosols is of 4 days period. Recent surface science studies of aerosols revealed that the

surfactants molecules can exist between the gas and particle interface of aerosols. It is reported that surfactants form a coating over particles and alter the cloud condensation activity by reducing the surface tension (McNeill et al., 2013; Ruehl et al., 2016). Cheng et al. (2019) have reported the existence of surfactants in the aerosol samples of marine atmosphere. They studied the interfacial behavior using Langmuir adsorption and the presence of surfactants was confirmed by IR spectroscopy. Vidović et al. (2020) confirmed the presence of brown carbon and SOA in the cloud droplets. A recent study conducted by Fankhauser et al. (2019) indicated the presence of bacterial traces in the aerosols over cloud droplets. The modelling studies revealed that these bacteria can degrade the complex organic compounds into fragments. However, their impacts could be less significant due to the presence of such bacteria in trace level. The physical separation of cloud droplets also limits the extent of bacterial degradation. Recently, Fe^{3+} and isoprene derived organosulfur SOA compounds have been found by Huang et al. (2019) in the atmospheric aerosols. Many research groups have also reported the presence of surface-active substances in the light absorbing compounds containing aerosols (Sareen et al., 2010; Schwie et al., 2010; Li et al., 2011). In another work, significant reduction in the interfacial tension of aerosols was reported due to photooxidation of brown carbons. Gubbins et al. (2019) investigated the influence of alcohol uptake onto organosulfur containing SOA system which revealed that alcohol uptake triggers light absorbing organic formation that leads to the increase in viscosity with increasing O:C ratio of the aerosols. Isoprene derived SOA formed via epoxydiols (IEPOX) causes high viscosity that reduces the further IEPOX uptake and results in formation of stable SOA (Olson et al., 2019; Zhang et al., 2019). All the recent advances open up new ways in atmospheric chemistry and aerosols dynamics. This would be helpful to fulfil the knowledge gaps and also in tackling atmospheric pollution.

7. Future Perspectives

Particles are formed and grow either by condensation or by partitioning on particles or by forming more volatile oxidation products. Hence, ELVOC need attention and analysis towards their formation mechanism and quantifications. The challenges as summarized could be- critical requirement of authentic quantitative standards those are currently unavailable, developing more understanding about the atmospheric chemistry of interface from micro to macro level in both gas to particle interface as well as interface between atmosphere and biosphere, cryosphere and hydrosphere, and advanced investigation on interaction of organic and inorganic components. Apropos to the above, advancement in accurate methodologies and high-resolution instruments are the needs of the hours. In this review, detailed mechanisms of BSOA formation is well discussed and besides these more studies are required to estimate the link between biogenic and anthropogenic primary emissions. Faster and accurate analytical techniques are further required for quantitative determination of BSOA—mainly their hourly and time series variation studies in the ambient atmosphere are required. The lifetime of BSOA, their distribution in different size ranges, their possibilities of long-range transport and their stability in the atmosphere are some challenges that should be dealt in future by researchers.

8. Conclusion

This review summarizes the different mechanism of gas phase oxidation, reactive uptake of formed gaseous species and subsequent biogenic SOA into aerosol particle surface. The significance of gaseous pollutants (NO_x , SO_2 , O_3) on the vegetation derived VOCs and their degradation under atmospheric conditions (temperature and relative humidity) are elaborated. On the basis of variability in the VOC emission rates the atmospheric chemistry are also different

during day and night time. The biogenic SOA formation and their seasonal variations depend on geographical differences and sites of unique atmospheric composition like urban atmosphere, rural regions, coastal areas, mount and forest sites. These regions have great importance because of different unique crossover between anthropogenic and biogenic pollutants. On the basis of the complexity and the optical properties of organic aerosols their environmental impacts have been discussed in terms of radiative forcing SOA. The widely used analytical techniques for estimation of organic aerosols have been summarized, and based on the information obtained by particular techniques; they are compared using 3dimensional representation onto chess-board scheme. For online measurement the AMS and for offline measurement 2D GC-MS have been considered to be the best suitable techniques for estimation of organic aerosols. Apart from the discussed analytical techniques there is still requirement of a technique with excellent chemical, time and size resolution, which could be able to estimate molecular identity, greater coverage of aerosols mass percent characterization for being able to be placed as near as possible toward the h8 position (represented as perfect instrument) of the chess board (described in results and discussion).

Declaration of Competing Interest

There is no conflict of interest.

Acknowledgement

Authors would like to acknowledge DST-FIST [No. SR/FST/CSI-259/2014(c)] and UGC-SAP [No. F-540/7/DRS-II/ 2016 (SAP-I)] for financial support. Mithlesh Mahilang is also thankful to Pt. Ravishankar Shukla University, Raipur, C.G., and India for providing university scholarship under the VR. No 1413/Fin/2016.

References

- Abbatt, J.P., Wang, C., 2020. The atmospheric chemistry of indoor environments. *Environ. Sci.: Processes Impacts* 2020, 22, 25-48.
- Agarwal, A., Satsangi, A., Lakhani, A., Kumari, K.M., 2020. Seasonal and spatial variability of secondary inorganic aerosols in PM_{2.5} at Agra: Source apportionment through receptor models. *Chemosphere* 242, 125-132.
- Aguilar-Dodier, L.C., Castillo, J.E., Quintana, P.J., Montoya, L.D., Molina, L.T., Zavala, M., Almanza-Veloz, V. and Rodríguez-Ventura, J.G., 2020. Spatial and temporal evaluation of H₂S, SO₂ and NH₃ concentrations near Cerro Prieto geothermal power plant in Mexico. *Atmos. Pollut. Res.*, 11(1), 94-104.
- Aiken, A. C., DeCarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Zhang, Q., Sun, Y., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., Jimenez, J. L., 2008. O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high resolution time-of-flight aerosol mass spectrometry. *Environ. Sci. Technol.* 42, 4478–4485.
- Allan, J., Morgan, W., Darbyshire, E., Flynn, M.J., Williams, P., Oram, D., Artaxo, P., Brito, J., Lee, J., Coe, H., 2014. Airborne observations of IEPOX-derived isoprene SOA in the Amazon during SAMBBA, *Atmos. Chem. Phys.*, 14, 11393–11407.
- Andreae, M. O., Crutzen, P. J., 1997. Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry. *Science* 276, 1052–1058.
- Atkinson, R., 2007. Rate constants for the atmospheric reactions of alkoxy radicals: a revised estimation method. *Atmos. Environ.* 41, 8468–8465.
- Atkinson, R., Arey, J., 2003. Gas-phase tropospheric chemistry of biogenic volatile organic compounds: a review. *Atmos. Environ.* 37, (2), 197–219.

- 788 Atkinson, R., Aschmann, S.M., 1993. Hydroxyl radical production from the gas-phase reactions
789 of ozone with a series of alkenes under atmospheric conditions. *Environ. Sci. Technol.*
790 27(7), 1357-1363.
- 791 Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin,
792 M. E., Rossi, M. J., Troe, J., 2006. IUPAC Subcommittee: Evaluated kinetic and
793 photochemical data for atmospheric chemistry: Volume II - gas phase reactions of
794 organic species. *Atmos. Chem. Phys.* 6, 3625–4055.
- 795 Babar, Z.B., Ashraf, F., Park, J.H., Lim, H.J., 2020. Volatility parameters of secondary organic
796 aerosol components determined using a thermal denuder. *Atmos. Environ.* 226, 117405.
- 797 Bagchi, A., Yu, Y., Huang, J.H., Tsai, C.C., Hu, W.P., Wang, C.C., 2020. Evidence and
798 evolution of Criegee intermediates, hydroperoxides and secondary organic aerosols
799 formed via ozonolysis of α -pinene. *Phys. Chem. Chem. Phys.* 22(12), 6528-6537.
- 800 Bates, K.H., Jacob, D.J., 2019. A new model mechanism for atmospheric oxidation of isoprene:
801 global effects on oxidants, nitrogen oxides, organic products, and secondary organic
802 aerosol. *Atmos. Chem. Phys.* 19(14), 9613-9640.
- 803 Baustian, K., Wise, M., Jensen, E., Schill, G., Freedman, M., Tolbert, M., 2013. State
804 transformations and ice nucleation in amorphous (semi-) solid organic aerosol, *Atmos.*
805 *Chem. Phys.* 13(11), 5615–5628.
- 806 Bhat, S., Fraser, M. P., 2007. Primary source attribution and analysis of α -pinene photooxidation
807 products in Duke Forest. North Carolina. *Atmos. Environ.* 41, 2958–2966.
- 808 Bianchi, F., Kurtén, T., Riva, M., Mohr, C., Rissanen, M.P., Roldin, P., Berndt, T., Crounse,
809 J.D., Wennberg, P.O., Mentel, T.F., Wildt, J., 2019. Highly oxygenated organic
810 molecules (HOM) from gas-phase autoxidation involving peroxy radicals: A key
811 contributor to atmospheric aerosol. *Chem. Rev.* 119 (6), 3472-3509.
- 812 Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., DeAngelo, B.J., Flanner,
813 M.G., Ghan, S., Karcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim,

- M.C., Schultz, M.G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P.K., Jacobson, M.Z., Kaiser, J.W., Klimont, Z., Lohmann, U., Schwarz, J.P., Shindell, D., Storelvmo, T., Warren, S.G., Zender, C.S., 2013. Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res.* 118, 5380–5552.
- Bonn, B., Kulmala, M., Riipinen, I., Sihto, S. L., Ruuskanen, T. M., 2008. How biogenic terpenes govern the correlation between sulfuric acid concentrations and new particle formation, *J. Geophys. Res.-Atmos.* 113, D12209.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., Zhang, X. Y. 2013. Clouds and Aerosols. In *Climate change 2013: the physical science basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, 571-657.
- Boy, M., Karl, T., Turnipseed, A., Mauldin, R. L., Kosciuch, E., Greenberg, J., Rathbone, J., Smith, J., Held, A., Barsanti, K., Wehner, B., Bauer, S., Wiedensohler, A., Bonn, B., Kulmala, M., Guenther, A., 2008. New particle formation in the Front Range of the Colorado Rocky Mountains, *Atmos. Chem. Phys.* 8, 1577– 1590.
- Boy, M., Thomson, E.S., Acosta Navarro, J.C., Arnalds, O., Batchvarova, E., Bäck, J., Berninger, F., Bilde, M., Brasseur, Z., Dagsson-Waldhauserova, P., Castarède, D., 2019. Interactions between the atmosphere, cryosphere, and ecosystems at northern high latitudes. *Atmos. Chem. Phys.* 19 (3), 2015-2061.
- Brüggemann, M., Xu, R., Tilgner, A., Kwong, K.C., Mutzel, A., Poon, H.Y., Otto, T., Schaefer, T., Poulain, L., Chan, M.N., Herrmann, H., 2020. Organosulfates in Ambient Aerosol: State of Knowledge and Future Research Directions on Formation, Abundance, Fate, and Importance. *Environ. Sci. Technol.* 54(7), 3767-3782.

- 839 Calfapietra, C., Fares, S., Manes, F., Morani, A., Sgrigna, G., Loreto, F., 2013. Role of Biogenic
 840 Volatile Organic Compounds (BVOC) emitted by urban trees on ozone concentration in
 841 cities: A review. *Environ. Pollut.* 183, 71-80.
- 842 Cantrell, W., Robinson, C., 2006. Heterogeneous freezing of ammonium sulfate and sodium
 843 chloride solutions by long chain alcohols, *Geophys. Res. Lett.* 33, L07802
- 844 Chen, L., Bao, Z., Wu, X., Li, K., Han, L., Zhao, X., Zhang, X., Wang, Z., Azzi, M., Cen, K.,
 845 2020. The effects of humidity and ammonia on the chemical composition of secondary
 846 aerosols from toluene/NO_x photo-oxidation. *Sci. Total Environ.* 728, 138671.
- 847 Cheng, S., Li, S., Tsona, N. T., George, C., Du, L., 2019. Insights into the Headgroup and Chain
 848 Length Dependence of Surface Characteristics of Organic-Coated Sea Spray Aerosols.
 849 *ACS Earth Space Chem.* 3 (4), 571–580.
- 850 Cheng, Y., He, K.B., Du, Z.Y., Zheng, M., Duan, F.K., Ma, Y.L., 2015. Humidity plays an
 851 important role in the PM_{2.5} pollution in Beijing. *Environ. Pollut.* 197, 68-75.
- 852 Choi, J.K., Ban, S.J., Kim, Y.P., Kim, Y.H., Yi, S.M., Zoh, K.D., 2015. Molecular marker
 853 characterization and source appointment of particulate matter and its organic aerosols.
 854 *Chemosphere*, 134, 482-491.
- 855 Ciccioli, P., Centritto, M., Loreto, F., 2014. Biogenic volatile organic compound emissions from
 856 vegetation fires. *Plant Cell Environ.* 37 (8), 1810-1825.
- 857 Claeys, M., Graham, B., Vas, G., Wang, W., Vermeylen, R., Pashynska, V., Cafmeyer, J.,
 858 Guyon, P., Andreae, M. O., Artaxo, P., Maenhaut, W., 2004. Formation of secondary
 859 organic aerosols through photooxidation of isoprene. *Science* 303, 1173–1176.
- 860 Clements, A. L., Seinfeld, J. H., 2007. Detection and quantification of 2-methyltetrols in ambient
 861 aerosol in the southeastern United States. *Atmos. Environ.* 41, 1825–1830.
- 862 Cui, T., Green, H.S., Selleck, P.W., Zhang, Z. O., Brien, R. E., Gold, A., Keywood, M., Kroll, J.
 863 H., Surratt, J.D., 2019. Chemical Characterization of Isoprene-and Monoterpene-

- Derived Secondary Organic Aerosol Tracers in Remote Marine Aerosols over a Quarter Century. *ACS Earth and Space Chem.* 3(6), 935-946.
- De Haan, D.O., Pajunoja, A., Hawkins, L.N., Welsh, H.G., Jimenez, N.G., De Loera, A., Zauscher, M.D., Andretta, A.D., Joyce, B.W., De Haan, A.C., Riva, M., 2019. Methylamine's effects on methylglyoxal-containing aerosol: chemical, physical, and optical changes. *ACS Earth and Space Chem.* 3(9), 1706-1716.
- Decesari, S., Mircea, M., Cavalli, F., Fuzzi, S., Moretti, F., Tagliavini, E., Facchini, M. C., 2007. Source attribution of watersoluble organic aerosol by nuclear magnetic resonance spectroscopy. *Environ. Sci. Technol.* 41, 2479–2484.
- Denkenberger, K. A., Moffet, R. C., Holecek, J. C., Rebotier, T. P., Prather, K. A., 2007. Real-time, single-particle measurements of oligomers in aged ambient aerosol particles. *Environ. Sci. Technol.* 41, 5439–5446.
- Deshmukh, D.K., Kawamura, K., Deb, M.K., 2016. Dicarboxylic acids, ω -oxocarboxylic acids, α -dicarbonyls, WSOC, OC, EC, and inorganic ions in wintertime size-segregated aerosols from central India: Sources and formation processes. *Chemosphere* 161, 27-42.
- Ding, X., Wang, X.M., Zheng, M., 2011. The influence of temperature and aerosol acidity on biogenic secondary organic aerosol tracers: Observations at a rural site in the central Pearl River Delta region, South China. *Atmos. Environ.* 45(6), 1303-1311.
- Dommen, J., Metzger, A., Duplissy, J., Kalberer, M., Alfarra, M. R., Gascho, A., Weingartner, E., Prévôt, A. S. H., Verheggen, B., Baltensperger, U., 2006. Laboratory observation of oligomers in the aerosol from isoprene/NO_x photooxidation. *Geophys. Res. Lett.* 31, L13805.
- Draper, D. C., Myllys, N., Hyttinen, N., Møller, K. H., Kjaergaard, H. G., Fry, J. L., Smith, J. N., Kurtén, T., 2019. Formation of Highly Oxidized Molecules from NO₃ Radical Initiated Oxidation of δ -3-Carene: A Mechanistic Study. *ACS Earth Space Chem.* 3 (8), 1460–1470.
- Du, C.Y., Yang, H., Wang, N., Pang, S.F., Zhang, Y.H., 2020. pH effect on the release of NH₃ from the internally mixed sodium succinate and ammonium sulfate aerosols. *Atmos. Environ.* 220, 117101.

- Edney, E. O., Kleindienst, T. E., Jaoui, M., Lewandowski, M., Offenberg, J. H., Wang, W.,
Claeys, M., 2005. Formation of 2-methyl tetrols and 2-methylglyceric acid in secondary
organic aerosol from laboratory irradiated isoprene/NO_x/SO₂/air mixtures and their
detection in ambient PM_{2.5} samples collected in the eastern United States. *Atmos.*
Environ. 39, 5281–5289.
- Ehn, M., Kleist, E., Junninen, H., Petaja, T., Lonn, G., Schobesberger, S., Dal Maso, M.,
Trimborn, A., Kulmala, M., Worsnop, D. R., Wahner, A., Wildt, J., Mentel, T. F., 2012.
Gas phase formation of extremely oxidized pinene reaction products in chamber and
ambient air. *Atmos. Chem. Phys.* 12 (11), 5113–5127.
- Ehn, M., Thornton, J. A., Kleist, E., Sipila, M., Junninen, H., Pullinen, I., Springer, M., Rubach,
F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I. H., Rissanen, M.,
Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurten,
T., Nielsen, L. B., Jorgensen, S., Kjaergaard, H. G., Canagaratna, M., Dal Maso, M.,
Berndt, T., Petaja, T., Wahner, A., Kerminen, V. M., Kulmala, M., Worsnop, D. R.,
Wildt, J., Mentel, T. F., 2014. A large source of low-volatility secondary organic
aerosol. *Nature* 506, 476–479.
- Faiola, C. L., Pullinen, I., Buchholz, A., Khalaj, F., Ylisirnio, A., Kari, E., Miettinen, P.,
Holopainen, J. K., Kivimäenpää, M., Schobesberger, S., Yli-Juuti, T., Virtanen, A.,
2019. Secondary Organic Aerosol Formation from Healthy and Aphid Stressed Scots
Pine Emissions. *ACS Earth Space Chem.* 3 (9), 1756–1772.
- Fairlie, T.D., Liu, H., Vernier, J.P., Campuzano-Jost, P., Jimenez, J.L., Jo, D.S., Zhang, B.,
Natarajan, M., Avery, M.A., Huey, G., 2020. Estimates of regional source contributions
to the Asian Tropopause Aerosol Layer using a chemical transport model. *J. Geophys.*
Res-Atmos. 125, e2019JD031506.

- 917 Fankhauser, A. M., Antonio, D. D., Krell, A., Alston, S. J., Banta, S., McNeill, V. F., 2019.
918 Constraining the Impact of Bacteria on the Aqueous Atmospheric Chemistry of Small
919 Organic Compounds. ACS Earth Space Chem. 3 (8), 1485–1491.
- 920 Fick, J., Pommer, L., Nilsson, C., Andersson, B., 2003. Effect of OH radicals, relative humidity,
921 and time on the composition of the products formed in the ozonolysis of α -pinene.
922 Atmos. Environ. 37, 4087-4096.
- 923 Finewax, Z., De Gouw, J. A., Ziemann, P. J., 2019. Products and Secondary Organic Aerosol
924 Yields from the OH and NO₃ Radical- Initiated Oxidation of Resorcinol. ACS Earth
925 Space Chem. 3 (7), 1248–1259.
- 926 Flores, R.M., Doskey, P.V., 2014. Using multidimensional gas chromatography to group
927 secondary organic aerosol species by functionality. Atmos. Environ. 96, 310-321.
- 928 Flores, R.M., Doskey, P.V., 2015. Evaluation of multistep derivatization methods for
929 identification and quantification of oxygenated species in organic aerosol. J. Chromatogr.
930 A 1418, 1-11.
- 931 Fu, P., Aggarwal, S.G., Chen, J., Li, J., Sun, Y., Wang, Z., Chen, H., Liao, H., Ding, A., Umarji,
932 G.S., Patil, R.S., 2016. Molecular markers of secondary organic aerosol in Mumbai,
933 India. Environ. Sci. Technol. 50(9), 4659-4667.
- 934 Fu, P., Kawamura, K., Pavuluri, C.M., Chen, J., Swaminathan, T., 2010. Contributions of
935 isoprene, α/β -pinene and β -caryophyllene to secondary organic aerosol in tropical
936 India. Low temperature science, 68, 79-88.
- 937 Garofalo, L. A., Pothier, M. A., Levin, E. J. T., Campos, T., Kreidenweis, S. M., Farmer, D. K.
938 Emission and Evolution of Submicron Organic Aerosol in Smoke from Wildfires in the
939 Western United States. ACS Earth Space Chem. 2019, 3 (7), 1237–1247.

- 940 Gen, M., Zhang, R., Huang, D., Li, Y., Chan, C.K.K., 2019. Heterogeneous Oxidation of SO₂ in
941 Sulfate Production During Nitrate Photolysis at 300 nm: Effect of pH, Relative
942 Humidity, Irradiation Intensity, and the Presence of Organic Compounds. *Environ. Sci.*
943 *Technol.* 53 (15), 8757-8766.
- 944 Geng, X., Mo, Y., Li, J., Zhong, G., Tang, J., Jiang, H., Ding, X., Malik, R.N., Zhang, G., 2020.
945 Source apportionment of water-soluble brown carbon in aerosols over the northern south
946 China sea: Influence from land outflow, SOA formation and marine emission. *Atmos.*
947 *Environ.* 229, 117484.
- 948 George, C., Ammann, M., D'Anna, B., Donaldson, D.J., Nizkorodov, S.A., 2015. Heterogeneous
949 photochemistry in the atmosphere. *Chem. Rev.* 115(10), 4218-4258.
- 950 Goldstein, A. H., Galbally, I. E., 2007. Known and unexplored organic constituents in the earth's
951 atmosphere. *Environ. Sci. Technol.* 41, 1514–1521.
- 952 Gorkowski, K., Donahue, N.M. and Sullivan, R.C., 2020. Aerosol Optical Tweezers Constrain
953 the Morphology Evolution of Liquid-Liquid Phase-Separated Atmospheric Particles.
954 *Chem* 6(1), 204-220.
- 955 Grace, D.N., Sharp, J.R., Holappa, R.E., Lugos, E.N., Sebold, M.B., Griffith, D.R., Hendrickson,
956 H.P., Galloway, M.M., 2019. Heterocyclic Product Formation in Aqueous Brown
957 Carbon Systems. *ACS Earth Space Chem.* 3(11), 2472-2481.
- 958 Gubbins, E. F., Miller, R., Inman, H., Bhat, T. A., Nwankwo, A. A. O., Myers, H., Berke, A. E.,
959 2019. Relationship between Structure, Functionality, and Viscosity for Aerosol-
960 Mimicking Solutions Containing Ammonium Sulfate, Glyoxal, and a Series of Oxidized
961 C1-C5 Compounds. *ACS Earth Space Chem.* 3 (8), 1492–1498.

- Gunsch, M. J., May, N. W., Wen, M., Bottenus, C. L. H., Gardner, D. J., VanReken, T. M., Bertman, S. B., Hopke, P. K., Ault, A. P., Pratt, K. A., 2018. Ubiquitous influence of wildfire emissions and secondary organic aerosol on summertime atmospheric aerosol in the forested Great Lakes region. *Atmos. Chem. Phys.* 18.5, 3701-3715.
- Guo, X., Ma, F., Liu, C., Niu, J., He, N., Chen, J., Xie, H.B., 2020. Atmospheric oxidation mechanism and kinetics of isoprene initiated by chlorine radicals: A computational study. *Sci. Total Environ.* 712, 136330.
- Hall, W. A., Johnston, M. V., 2011. Oligomer Content of alpha-Pinene Secondary Organic Aerosol, *Aerosol Sci. Technol.* 45, 37–45.
- Hallquist, M., Wenger, J.C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N.M., George, C., Goldstein, A.H., Hamilton, J.F., 2009. The formation, properties and impact of secondary organic aerosol: current and emerging issues. *Atmos. Chem. Phys.* 9(14), 5155-5236.
- Hamilton, J. F., Webb, P. J., Lewis, A. C., Hopkins, J. R., Smith, S., Davy, P., 2004. Partially oxidised organic components in urban aerosol using GCXGC-TOF/MS. *Atmos. Chem. Phys.* 4, 1279– 1290.
- Haque, M.M., Kawamura, K. and Kim, Y., 2016. Seasonal variations of biogenic secondary organic aerosol tracers in ambient aerosols from Alaska. *Atmos. Environ.* 130, 95-104.
- Harley, P.C., Monson, R.K., Lerdau, M.T., 1999. Ecological and evolutionary aspects of isoprene emission from plants. *Oecologia* 118, 109–123.
- Haywood, J., Boucher, O., 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review. *Rev. Geophys.* 38 (4), 513–543.
- Healy, R. M., Wenger, J. C., Metzger, A., Duplissy, J., Kalberer, M., Dommen, J., 2008. Gas/particle partitioning of carbonyls in the photooxidation of isoprene and 1,3,5-trimethylbenzene. *Atmos. Chem. Phys.* 8, 3215–3230.

- 987 Helmig, D., Daly, R.W., Milford, J., Guenther, A., 2013. Seasonal trends of biogenic terpene
988 emissions. *Chemosphere* 93(1), 35-46.
- 989 Hewitt, C. N., Lee, J. D., MacKenzie, A. R., Barkley, M. P., Carslaw, N., Carver, G. D.,
990 Chappell, N. A., Coe, H., Collier, C., Commane, R., Davies, F., Davison, B., DiCarlo, P.,
991 Di Marco, C. F., Dorsey, J. R., Edwards, P. M., Evans, M. J., Fowler, D., Furneaux, K.
992 L., Gallagher, M., Guenther, A., Heard, D. E., Helfter, C., Hopkins, J., Ingham, T., Irwin,
993 M., Jones, C., Karunaharan, A., Langford, B., Lewis, A. C., Lim, S. F., MacDonald, S.
994 M., Mahajan, A. S., Malpass, S., Mc-Figgans, G., Mills, G., Misztal, P., Moller, S.,
995 Monks, P. S., Nemitz, E., Nicolas-Perea, V., Oetjen, H., Oram, D. E., Palmer, P. I.,
996 Phillips, G. J., Pike, R., Plane, J. M. C., Pugh, T., Pyle, J. A., Reeves, C. E., Robinson, N.
997 H., Stewart, D., Stone, D., Whalley, L. K., and Yin, X., 2010. Overview: oxidant and
998 particle photochemical processes above a south-east Asian tropical rainforest (the OP3
999 project): introduction, rationale, location characteristics and tools, *Atmos. Chem. Phys.*,
1000 10, 169–199.
- 1001 Hinks, M. L., M. V., Brady, H., Lignell, M. J., Song, J. W., Grayson, A. K., Bertram, P., Lin, A.,
1002 Laskin, J., Nizkorodov, S. A., 2016. Effect of viscosity on photodegradation rates in
1003 complex secondary organic aerosol materials. *Phys. Chem. Chem. Phys.* 18 (13), 8785–
1004 8793.
- 1005 Hong, Z., Zhang, H., Zhang, Y., Xu, L., Liu, T., Xiao, H., Hong, Y., Chen, J., Li, M., Deng, J.,
1006 Wu, X., 2019. Secondary organic aerosol of PM_{2.5} in a mountainous forest area in
1007 southeastern China: Molecular compositions and tracers implication. *Sci. Total Environ.*
1008 653, 496-503.
- 1009 Hoyle, C. R., Boy, M., Donahue, N. M., Fry, J. L., Glasius, M., Guenther, A., Hallar, A. G.,
1010 Hartz, K. H., Petters, M. D., Petaja, T., Rosenoern, T., Sullivan, A. P., 2011. A review
1011 of the anthropogenic influence on biogenic secondary organic aerosol. *Atmos. Chem.*
1012 *Phys.* 11 (1), 321–343.
- 1013 Hoyle, C. R., Boy, M., Donahue, N. M., Fry, J. L., Glasius, M., Guenther, A., Hallar, A. G., Huff
1014 Hartz, K., Petters, M. D., Petäjä, T., Rosenoern, T., Sullivan, A. P., 2009. The
1015 formation, properties and impact of secondary organic aerosol: current and emerging
1016 issues. *Atmos. Chem. Phys.* 9, 5155–5236.

- 1017 Huang, L., Coddens, E. M., Grassian, V. H., 2019. Formation of Organosulfur Compounds from
1018 Aqueous Phase Reactions of S(IV) with Methacrolein and Methyl Vinyl Ketone in the
1019 Presence of Transition Metal Ions. *ACS Earth Space Chem.* 3 (9), 1749– 1755.
- 1020 Ide, Y., Uchida, K., Takegawa, N., 2019. Ionization efficiency of evolved gas molecules from
1021 aerosol particles in a thermal desorption aerosol mass spectrometer: Numerical
1022 simulations. *Aerosol Sci. Tech.* 53, 1-10.
- 1023 Iinuma, Y., Müller, C., Böge, O., Gnauk, T., Herrmann, H., 2007. The formation of organic
1024 sulfate esters in the limonene ozonolysis secondary organic aerosol (SOA) under acidic
1025 conditions. *Atmos. Environ.* 41, 5571–5583.
- 1026 Ion, A. C., Vermeylen, R., Kourtchev, I., Cafmeyer, J., Chi, X., Gelencsér, A., Maenhaut, W.,
1027 Claeys, M., 2005. Polar organic compounds in rural PM_{2.5} aerosols from K-puszt, Hungary, during a 2003 summer field campaign: sources and diurnal variations. *Atmos. Chem. Phys.* 5, 1805–1814.
- 1030 Isaacman, G., Kreisberg, N. M., Yee, L. D., Worton, D. R., Chan, A. W. H., Moss, J. A., Hering,
1031 S. V., Goldstein, A. H., 2014. Online derivatization for hourly measurements of gas-
1032 and particle-phase semi-volatile oxygenated organic compounds by thermal desorption
1033 aerosol gas chromatography (SV-TAG). *Atmos. Meas. Tech.* 7 (12), 4417–4429.
- 1034 Izhar, S., Gupta, T., Panday, A.K., 2020. Scavenging efficiency of water soluble inorganic and
1035 organic aerosols by fog droplets in the Indo Gangetic Plain. *Atmos. Res.* 235, 104767.
- 1036 Jang, M., Czoschke, N.M., Lee, S., Kamens, R.M., 2002. Heterogeneous atmospheric aerosol
1037 production by acid-catalyzed particle-phase reactions. *Science* 298 (5594), 814-817.
- 1038 Jang, M., Sun, S., Winslow, R., Han, S., Yu, Z., 2020. In Situ Aerosol Acidity Measurements
1039 Using a UV-Visible Micro-Spectrometer and its Application to the Ambient Air. *Aerosol*
1040 *Sci. Tech.* 1-30.
- 1041 Jaoui, M., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., Edney, E. O., 2005.
1042 Identification and quantification of aerosol polar oxygenated compounds bearing

- 1043 carboxylic or hydroxyl groups: 2. Organic tracer compounds from monoterpenes.
1044 Environ. Sci. Technol. 39, 5661–5673.
- 1045 Jaoui, M., Kleindienst, T.E., Lewandowski, M., Edney, E.O., 2004. Identification and
1046 quantification of aerosol polar oxygenated compounds bearing carboxylic and/or
1047 hydroxyl groups. 1. Method development. Anal. Chem. 76, 4765-4778.
- 1048 Johnson, D., Marston, G., 2008. The gas-phase ozonolysis of unsaturated volatile organic
1049 compounds in the troposphere. Chem. Soc. Rev. 37, 699–716.
- 1050 Johnston, M.V., Kerecman, D.E., 2019. Molecular Characterization of Atmospheric Organic
1051 Aerosol by Mass Spectrometry. Annu. Rev. Anal. Chem. 12, 247-274.
- 1052 Jokinen, T., Sipila, M., Richters, S., Kerminen, V. M., Paasonen, P., Stratmann, F., Worsnop, D.
1053 R., Kulmala, M., Ehn, M., Herrmann, H., Berndt, T., 2014. Rapid autoxidation forming
1054 highly oxidized RO₂ radicals in the atmosphere. Angew. Chem., Int. Ed. 125 (25),
1055 14825–14829.
- 1056 Kamens, R. M., Jang, M., Chien, C. J., Leach, K. Aerosol formation from the reaction of α -
1057 pinene and ozone using a gas-phase kinetics-aerosol partitioning model. Environ. Sci.
1058 Technol. 1999, 33, 1430-1438.
- 1059 Kammer, J., Flaud, P.M., Chazeaubeny, A., Ciuraru, R., Le Menach, K., Geneste, E., Budzinski,
1060 H., Bonnefond, J.M., Lamaud, E., Perraudin, E., Villenave, E., 2020. Biogenic volatile
1061 organic compounds (BVOCs) reactivity related to new particle formation (NPF) over the
1062 Landes forest. Atmos. Res. 237, 104869.
- 1063 Kang, M., Fu, P., Kawamura, K., Yang, F., Zhang, H., Zang, Z., Ren, H., Ren, L., Zhao, Y., Sun,
1064 Y., Wang, Z., 2018. Characterization of biogenic primary and secondary organic
1065 aerosols in the marine atmosphere over the East China Sea. Atmos. Chem. Phys.
1066 18(19), 13947-13967.

- 1067 Kawamura, K., Hoque, M. M. M., Bates, T. S., Quinn, P.K., 2017. Molecular distributions and
1068 isotopic compositions of organic aerosols over the western North Atlantic: Dicarboxylic
1069 acids, related compounds, sugars, and secondary organic aerosol tracers. *Org. Geochem.*
1070 113, 229-238.
- 1071 Kelly, J.M., Doherty, R.M., O'Connor, F.M., Mann, G.W., 2018. The impact of biogenic,
1072 anthropogenic, and biomass burning volatile organic compound emissions on regional
1073 and seasonal variations in secondary organic aerosol. *Atmos. Chem. Phys.* 18(10), 7393-
1074 7422.
- 1075 Kleindienst, T.E., Lewandowski, M., Offenberg, J.H., Jaoui, M., Edney, E.O., 2009. Formation
1076 of secondary organic aerosol from the isoprene β OH reaction in the absence of NO_x.
1077 *Atmos. Chem. Phys.*, 9, 6541–6558.
- 1078 Kołodziejczyk, A., Pyrcz, P., Błaziak, K., Pobudkowska, A., Sarang, K., Szmigielski, R., 2020.
1079 Physicochemical Properties of Terebic Acid, MBTCA, Diaterpenylic Acid Acetate, and
1080 Pinanediol as Relevant α -Pinene Oxidation Products. *ACS Omega*, 5, 14, 7919-7927.
- 1081 Koop, T., Bookhold, J., Shiraiwa, M., Pöschl, U. 2011. Glass transition and phase state of
1082 organic compounds: Dependency on molecular properties and implications for secondary
1083 organic aerosols in the atmosphere, *Phys. Chem. Chem. Phys.*, 13(43), 19238-19255.
- 1084 Kotianova, P., Matisova, E., Puxbaum, H., Lehotay, J., 2004. Analysis of organic compounds in
1085 aerosols. A review. *Chem. Anal. (Warsaw)* 49, 833–844.
- 1086 Kourtchev, I., Ruuskanen, T., Maenhaut, W., Kulmala, M., Claeys, M., 2005. Observation of 2-
1087 methyltetrols and related photo-oxidation products of isoprene in boreal forest aerosols
1088 from Hyytiälä, Finland. *Atmos. Chem. Phys. Discuss.*, 5, 2947–2971.

- 1089 Kourtchev, I., Warnke, J., Maenhaut, W., Hoffmann, T., Claeys, M., 2008. Polar organic marker
1090 compounds in PM_{2.5} aerosol from a mixed forest site in western Germany. *Chemosphere*,
1091 73(8), 1308-1314.
- 1092 Kulmala, M., Toivonen, A., Makela, J. M., Laaksonen, A., 1998. Analysis of the growth of
1093 nucleation mode particles observed in Boreal forest. *Tellus Ser. B* 50 (5), 449–462
- 1094 Ladino, L. A., S. Zhou, J. D. Yakobi-Hancock, D. Aljawhary, P. D. Abbatt, 2014. Factors
1095 controlling the ice nucleating abilities of α -pinene SOA particles, *J. Geophys. Res.*
1096 *Atmos.* 119, 9041–9051.
- 1097 Lechevallier, L., Grilli, R., Kerstel, E., Romanini, D., Chappellaz, J., 2019. Simultaneous
1098 detection of C₂H₆, CH₄, and δ 13C-CH₄ using optical feedback cavity-enhanced
1099 absorption spectroscopy in the mid-infrared region: towards application for dissolved
1100 gas measurements. *Atmos. Meas. Tech. Discuss.* 12 (6), 3101-3109.
- 1101 Lee, L., Teng, A. P., Wennberg, P. O., Crounse, J. D., Cohen, R. C., 2014. On rates and
1102 mechanisms of OH and O₃ reactions with isoprene-derived hydroxy nitrates. *J. Phys.*
1103 *Chem. A*, 118, 1622– 1637.
- 1104 Lengyel, J., Pysanenko, A., Fárniková, K., Pluhařová, E. and Fárník, M., 2020. Oxidation
1105 Enhances Aerosol Nucleation: Measurement of Kinetic Pickup Probability of Organic
1106 Molecules on Hydrated Acid Clusters. *J. Phys. Chem. Lett.* 11(6), 2101-2105.
- 1107 Li, G., Cheng, Y., Kuhn, U., Xu, R., Yang, Y., Meusel, H., Wang, Z., Ma, N., Wu, Y., Li, M.,
1108 Williams, J., 2019. Physicochemical uptake and release of volatile organic compounds
1109 by soil in coated-wall flow tube experiments with ambient air. *Atmos. Chem. Phys.* 19
1110 (4), 2209-2232.
- 1111 Li, H., Ning, A., Zhong, J., Zhang, H., Liu, L., Zhang, Y., Zhang, X., Zeng, X.C., He, H., 2020.
1112 Influence of atmospheric conditions on sulfuric acid-dimethylamine-ammonia-based new
1113 particle formation. *Chemosphere*, 245, 25554.

- 1114 Li, J.J., Wang, G.H., Cao, J.J., Wang, X.M., Zhang, R.J., 2013. Observation of biogenic
1115 secondary organic aerosols in the atmosphere of a mountain site in central China:
1116 temperature and relative humidity effects. *Atmos. Chem. Phys.* 13, 11535-11549.
- 1117 Li, Z., Schwier, A. N., Sareen, N., McNeill, V. F., 2011. Reactive Processing of Formaldehyde
1118 and Acetaldehyde in Aqueous Aerosol Mimics: Surface Tension Depression and
1119 Secondary Organic Products. *Atmos. Chem. Phys.* 11 (22), 11617–11629.
- 1120 Liao, H., Henze, D.K., Seinfeld, J.H., Wu, S., Mickley, L.J., 2007. Biogenic secondary organic
1121 aerosol over the United States: comparison of climatological simulations with
1122 observations, *J. Geophys. Res.* 112, D06201.
- 1123 Lim, C.Y., Hagan, D.H., Coggon, M.M., Koss, A.R., Sekimoto, K., Gouw, J.D., Warneke, C.,
1124 Cappa, C.D., Kroll, J.H., 2019. Secondary organic aerosol formation from the laboratory
1125 oxidation of biomass burning emissions. *Atmos. Chem. Phys.* 19(19), 12797-12809.
- 1126 Lin, G., Sillman, S., Penner, J.E., Ito, A., 2014. Global modeling of SOA: the use of different
1127 mechanisms for aqueous-phase formation. *Atmos. Chem. Phys.* 14(11) 5451.
- 1128 Lin, P. J., Liu, J. E., Shilling, S. M., Kathmann, J., Laskin, A., 2015. Molecular characterization
1129 of brown carbon (BrC) chromophores in secondary organic aerosol generated from
1130 photo-oxidation of toluene. *Phys. Chem. Chem. Phys.* 17 (36), 312–325.
- 1131 Link, M.F., Nguyen, T.B., Bates, K.H., Müller, J.F., Farmer, D.K., 2020. Can isoprene oxidation
1132 explain high concentrations of atmospheric formic and acetic acid over forests?. *ACS*
1133 *Earth Space Chem.* <https://doi.org/10.1021/acsearthspacechem.0c00010>.
- 1134 Liu, P. F. N., Abdelmalki, H. M., Hung, Y., Wang, W., Brune, H., Martin, S. T., 2015.
1135 Ultraviolet and visible complex refractive indices of secondary organic material
1136 produced by photooxidation of the aromatic compounds' toluene and m-xylene. *Atmos.*
1137 *Chem. Phys.* 15 (3), 1435–1446.

- 1138 Liu, Q., Liu, D., Gao, Q., Tian, P., Wang, F., Zhao, D., Bi, K., Wu, Y., Ding, S., Hu, K., Zhang,
1139 J., 2020. Vertical characteristics of aerosol hygroscopicity and impacts on optical
1140 properties over the North China Plain during winter. *Atmos. Chem. Phys.* 20(6), 3931-
1141 3944.
- 1142 Liu, S., Jiang, X., Tsona, N.T., Lv, C., Du, L., 2019b. Effects of NO_x, SO₂ and RH on the SOA
1143 formation from cyclohexene photooxidation. *Chemosphere* 216, 794-804.
- 1144 Liu, Y., Li, X., Tang, G., Wang, L., Lv, B., Guo, X., Wang, Y., 2019a. Secondary organic
1145 aerosols in Jinan, an urban site in North China: Significant anthropogenic contributions
1146 to heavy pollution. *Int. J. Environ. Sci.* 80, 107-115.
- 1147 Mahilang, M., Deb, M.K., 2020. Seasonal variation and health implications of long-range
1148 transported and provincial size distributed aerosols at eastern central India. *J. Indian*
1149 *Chem. Soc.* 97, 85-100.
- 1150 Mahilang, M., Deb, M.K., Nirmalkar, J., Pervez, S., 2020. Influence of fireworks emission on
1151 aerosol aging process at lower troposphere and associated health risks in an urban region
1152 of eastern central India. *Atmos. Pollut. Res.* 11 (2020) 1127–1141.
- 1153 Majumdar, D., Rajaram, B., Meshram, S. and Chalapati Rao, C.V., 2012. PAHs in road dust:
1154 ubiquity, fate, and summary of available data. *Crit. Rev. Env. Sci. Tec.* 42(12), 1191-
1155 1232.
- 1156 Malavelle, F. F., Haywood, J. M., Mercado, L. M., Folberth, G. A., Bellouin, N., Sitch, S.,
1157 Artaxo, P., 2019. Studying the impact of biomass burning aerosol radiative and climate
1158 effects on the Amazon rainforest productivity with an Earth system model. *Atmos.*
1159 *Chem. Phys.* 19 (2), 1301-1326.
- 1160 Marais, E. A., Daniel J. J., Jose L. J., Pedro C. J., Douglas A. D., 2016. Aqueous-phase
1161 mechanism for secondary organic aerosol formation from isoprene: application to the
1162 southeast United States and co-benefit of SO₂ emission controls. *Atmos. Chem. Phys.*
1163 16 (3), 1603-18.

- 1164 Mauree, D., Naboni, E., Cocco, S., Perera, A.T.D., Nik, V.M., Scartezzini, J.L., 2019. A
1165 review of assessment methods for the urban environment and its energy sustainability to
1166 guarantee climate adaptation of future cities. *Renew. Sust. Energ. Rev.* 112, 733-746.
- 1167 McNeill, V. F., Sareen, N., Schwier, A. N., 2013. Surface-Active Organics in Atmospheric
1168 Aerosols. *Top. Curr. Chem.* 339, 201– 259.
- 1169 Mekic, M., Liu, J., Zhou, W., Loisel, G., Cai, J., He, T., Jiang, B., Yu, Z., Lazarou, Y.G., Li, X.
1170 and Brigante, M., 2019. Formation of highly oxygenated multifunctional compounds
1171 from cross-reactions of carbonyl compounds in the atmospheric aqueous phase.
1172 *Atmos. Environ.* 219, 117046.
- 1173 Mentel, T. F., Springer, M., Ehn, M., Kleist, E., Pullinen, I., Kurten, T., Rissanen, M., Wahner,
1174 A., Wildt, J., 2015. Formation of highly oxidized multifunctional compounds:
1175 autoxidation of peroxy radicals formed in the ozonolysis of alkenes – deduced from
1176 structure–product relationships. *Atmos. Chem. Phys.* 15, 6745–6765.
- 1177 Moffet, R. C., Prather, K. A., 2009. In-situ measurements of the mixing state and optical
1178 properties of soot with implications for radiative forcing estimates. *P. Natl. Acad. Sci.*
1179 *USA*, 106, 11872– 11877.
- 1180 Mohler, O., S. Benz, H. Saathoff, M. Schnaiter, R. Wagner, J. Schneider, S. Walter, V. Ebert, S.
1181 Wagner, 2008. The effect of organic coating on the heterogeneous ice nucleation
1182 efficiency of mineral dust aerosols, *Environ. Res. Lett.*, 3(2), 025007.
- 1183 Moise, T., Flores, J. M., Rudich Y., 2015. Optical properties of secondary organic aerosols and
1184 their changes by chemical processes. *Chem. Rev.* 115 (10), 4400–4439.
- 1185 Murray, B.J., Wilson, T.W., Dobbie, S., Cui, Z., Al-Jumur, S.M., Möhler, O., Schnaiter, M.,
1186 Wagner, R., Benz, S., Niemand, M., Saathoff, H., 2010. Heterogeneous nucleation of ice
1187 particles on glassy aerosols under cirrus conditions, *Nat. Geosci.* 3(4), 233–237.

- 1188 Mutzel, A., Poulain, L., Berndt, T., Iinuma, Y., Rodigast, M., Böge, O., Richters, S., Spindler,
 1189 G., Sipilä, M., Jokinen, T., Kulmala, M., 2015. Highly oxidized multifunctional organic
 1190 compounds observed in tropospheric particles: A field and laboratory study. *Environ. Sci.*
 1191 *Technol.* 49(13), 7754-7761.
- 1192 Myhre, G., Bjørn H. S., Schulz, M., Balkanski, Y., Bauer, S., Terje, K. B., Huisheng, B., Nicolas
 1193 B., Chin, M., Diehl, T., Easter, R. C., Johann F., Ghan, S. J., Hauglustaine, D., Trond I.,
 1194 Stefan K., Kirkevåg, A., Lamarque, J. F., Lin, G., Xiaohong L., Marianne T. L., Luo,
 1195 G., Ma, X., Van T., Penner, J. E., Rasch, P. J., Ruiz, A., Ragnhild, B. S., Stier, P.,
 1196 Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu, H., Yu, F., Yoon, J. H.,
 1197 Zhang, K., Zhang, H., Zhouet, C., 2013. Radiative forcing of the direct aerosol effect
 1198 from AeroCom Phase II simulations. *Atmos. Chem. Phys.* 13 (4), 1853–1877.
- 1199 Ng, N. L., Kwan, A. J., Surratt, J. D., Chan, A. W. H., Chhabra, P. S., Sorooshian, A., Pye, H. O.
 1200 T., Crounse, J. D., Wennberg, P. O., Flagan, R. C., Seinfeld, J. H., 2008. Secondary
 1201 organic aerosol (SOA) formation from reaction of isoprene with nitrate radicals (NO₃).
 1202 *Atmos. Chem. Phys.* 8, 4117–4140.
- 1203 Ng, N.L., Kroll, J.H., Keywood, M.D., Bahreini, R., Varutbangkul, V., Flagan, R.C., Seinfeld,
 1204 J.H., Lee, A. and Goldstein, A.H., 2006. Contribution of first-versus second-generation
 1205 products to secondary organic aerosols formed in the oxidation of biogenic hydrocarbons.
 1206 *Environ. Sci. Technol.* 40(7), 2283-2297.
- 1207 Nguyen, T. B., Crounse, J. D., Teng, A. P., St. Clair, J. M., Paulot, F., Wolfe, G. M., Wennberg,
 1208 P. O., 2015. Rapid deposition of oxidized biogenic compounds to a temperate forest. *P.*
 1209 *Natl. Acad. Sci. USA*, 112, 392–401.
- 1210 Novakov, T., Penner, J.E., 1993. Large contribution of organic aerosols to cloud-condensation-
 1211 nuclei concentrations. *Nature*, 365(6449), 823-826.
- 1212 Nozière, B., Kalberer, M., Claeys, M., Allan, J., D'Anna, B., Decesari, S., Finessi, E., Glasius,
 1213 M., Grgić, I., Hamilton, J. F., Hoffmann, T., Iinuma, Y., Jaoui, M., Kahnt, A., Kampf,

- 1214 C. J., Kourtchev, I., Maenhaut, W., Marsden, N., Saarikoski, S., Schnelle- Kreis, J.,
1215 Surratt, J. D., Szidat, S., Szmigielski, R., Wisthaler, A., 2015. The Molecular
1216 Identification of Organic Compounds in the Atmosphere: State of the Art and
1217 Challenges. *Chem. Rev.* 115 (10), 3919– 3983.
- 1218 Oberdorster, G., Oberdorster, E., Oberdorster, J., 2005. Nanotoxicology: An emerging discipline
1219 evolving from studies of ultrafine particles. *Environ. Health Persp.* 113, 823–839.
- 1220 Offenberg, J.H., Lewandowski, M., Edney, E.O., Kleindienst, T.E., Jaoui, M., 2009. Influence of
1221 aerosol acidity on the formation of secondary organic aerosol from biogenic precursor
1222 hydrocarbons. *Environ. Sci. Technol.* 43, 7742-7747.
- 1223 Olson, N. E., Lei, Z., Craig, R. L., Zhang, Y., Chen, Y., Lambe, A. T., Zhang, Z., Gold, A.,
1224 Surratt, J. D., Ault, A. P., 2019. Reactive Uptake of Isoprene Epoxydiols Increases the
1225 Viscosity of the Core of Phase- Separated Aerosol Particles. *ACS Earth Space Chem.* 3
1226 (8), 1402–1414.
- 1227 Padhy, P.K., Varshney, C.K., 2005. Emission of volatile organic compounds (VOC) from
1228 tropical plant species in India. *Chemosphere* 59(11), 1643-1653.
- 1229 Pagonis, D., Price, D., Algrim, L.B., Day, D.A., Handschy, A., Stark, H., Miller, S.L., de Gouw,
1230 J.A., Jimenez, J.L., Ziemann, P.J., 2019. Time-Resolved Measurements of Indoor
1231 Chemical Emissions, Deposition, and Reactions in a University Art Museum. *Environ.*
1232 *Sci. Technol.* 53 (9), 4794-4802.
- 1233 Pandey, A., Hsu, A., Tiwari, S., Pervez, S., Chakrabarty, R.K., 2020. Light absorption by organic
1234 aerosol emissions rivals that of black carbon from residential biomass fuels in South
1235 Asia. *Environ. Sci. Technol. Lett.* 7(4), 266-272.
- 1236 Pandey, A., Pervez, S., Chakrabarty, R.K., 2018. Aerosol light absorption and associated
1237 warming impacts from cookstove emissions in India. *AGUFM*, A41G-3022.

- 1238 Pani, S.K., Chantara, S., Khamkaew, C., Lee, C.T., Lin, N.H., 2019. Biomass burning in the
 1239 northern peninsular Southeast Asia: Aerosol chemical profile and potential
 1240 exposure. *Atmos. Res.* 224,180-195.
- 1241 Pani, S.K., Lin, N.H., Chantara, S., Wang, S.H., Khamkaew, C., Prapamontol, T., Janjai, S.,
 1242 2018. Radiative response of biomass-burning aerosols over an urban atmosphere in
 1243 northern peninsular Southeast Asia *Sci. Total Environ.* 633, 892-911.
- 1244 Pani, S.K., Wang, S.H., Lin, N.H., Tsay, S.C., Lolli, S., Chuang, M.T., Lee, C.T., Chantara, S.,
 1245 Yu, J.Y., 2016. Assessment of aerosol optical property and radiative effect for the layer
 1246 decoupling cases over the northern South China Sea during the 7th SEAS/Dongsha
 1247 Experiment. *J. Geophys. Res. Atmos.* 121(9), 4894-4906.
- 1248 Popovicheva, O.B., Engling, G., Diapouli, E., Saraga, D., Persiantseva, N.M., Timofeev, M.A.,
 1249 Kireeva, E.D., Shonija, N.K., Chen, S.H., Nguyen, D.L., Eleftheriadis, K., 2016. Impact
 1250 of smoke intensity on size-resolved aerosol composition and microstructure during the
 1251 biomass burning season in Northwest Vietnam. *Aerosol Air Qual. Res.* 16(11), 2635-
 1252 2654.
- 1253 Poschl, U., 2005. Atmospheric Aerosols: Composition, Transformation, Climate and Health
 1254 Effects. *Angew. Chem. Int. Edit.* 44, 7520– 7540.
- 1255 Pratap, V., Battaglia, M.A., Carlton, A.G., Hennigan, C.J., 2020. No evidence for brown carbon
 1256 formation in ambient particles undergoing atmospherically relevant drying. *Environ. Sci.*
 1257 *Process Impacts* 22(2), 442-450.

- 1258 Prenni, A. J., Petters, M. D., Faulhaber, A., Carrico, Ch. M., Ziemann, P. J., Kreidenweis, S. M.,
 1259 DeMott, P. J., 2009. Heterogeneous ice nucleation measurements of secondary organic
 1260 aerosol generated from ozonolysis of alkenes, *Geophys. Res. Lett.*, 36, L06808.
- 1261 Qi, X., Zhu, S., Zhu, C., Hu, J., Lou, S., Xu, L., Dong, J., Cheng, P., 2020. Smog chamber study
 1262 of the effects of NO_x and NH₃ on the formation of secondary organic aerosols and
 1263 optical properties from photo-oxidation of toluene. *Sci. Total Environ.* 727,138632.
- 1264 Qian, Y., Deng, G.H., Lapp, J. and Rao, Y., 2019. Interfaces of Gas–Aerosol Particles: Relative
 1265 Humidity and Salt Concentration Effects. *J. Phys. Chem. A* 123(29), 6304-6312.
- 1266 Quante, M., 2004, The role of clouds in the climate system, *J. Phys. IV France*, 121, 61–86
- 1267 Ram, K., Sarin, M.M., Tripathi, S.N., 2012. Temporal trends in atmospheric PM_{2.5}, PM₁₀,
 1268 elemental carbon, organic carbon, water-soluble organic carbon, and optical properties:
 1269 impact of biomass burning emissions in the Indo-Gangetic Plain. *Environ. Sci. technol.*
 1270 46 (2), 686-695.
- 1271 Ramanathan, V.C.P.J., Crutzen, P.J., Kiehl, J.T., Rosenfeld, D., 2001. Aerosols, climate, and the
 1272 hydrological cycle. *Science*, 294 (5549), 2119-2124.
- 1273 Ren, Y., Wang, G., Li, J., Wu, C., Cao, C., Wang, J., Zhang, L., Meng, F., Li, H., 2018. Seasonal
 1274 variation and size distribution of biogenic secondary organic aerosols at urban and
 1275 continental background sites of China. *J. Environ. Sci.* 71, 32-44.
- 1276 Ren, Y., Wang, G., Tao, J., Zhang, Z., Wu, C., Wang, J., Li, J., Wei, J., Li, H., Meng, F., 2019.
 1277 Seasonal characteristics of biogenic secondary organic aerosols at Mt. Wuyi in
 1278 Southeastern China: Influence of anthropogenic pollutants. *Environ. Pollut.* 252, 493-
 1279 500.
- 1280 Riemer, N., West, M., 2013. Quantifying aerosol mixing state with entropy and diversity
 1281 measures. *Atmos. Chem. Phys.* 13, 11423– 11439.

- 1282 Rissanen, T., Hyötyläinen, T., Kallio, M., Kronholm, J., Kulmala, M., Riekkola, M.L., 2006.
1283 Characterization of organic compounds in aerosol particles from a coniferous forest by
1284 GC–MS. *Chemosphere*, 64(7), 1185–1195.
- 1285 Ruehl, C. R., Davies, J. F., Wilson, K. R. An Interfacial Mechanism for Cloud Droplet
1286 Formation on Organic Aerosols. *Science* 2016, 351 (6280), 1447–1450.
- 1287 Salcedo, D., Onasch, T. B., Dzepina, K., Canagaratna, M. R., Zhang, Q., Huffman, J. A.,
1288 DeCarlo, P. F., Jayne, J. T., Mortimer, P., Worsnop, D. R., Kolb, C. E., Johnson, K. S.,
1289 Zuberi, B., Marr, L. C., Volkamer, R., Molina, L. T., Molina, M. J., Cardenas, B.,
1290 Bernabé, R. M., Márquez, C., Gaffney, J. S., Marley, N. A., Laskin, A.,
1291 Shutthanandan, V., Xie, Y., Brune, W., Leshner, R., Shirley, T., Jimenez, J. L., 2006.
1292 Characterization of ambient aerosols in Mexico City during the MCMA-2003 campaign
1293 with Aerosol Mass Spectrometry: results from the CENICA Supersite. *Atmos. Chem.*
1294 *Phys.* 6, 925–946.
- 1295 Sareen, N., Schwier, A. N., Shapiro, E. L., Mitroo, D., McNeill, V. F., 2010. Secondary Organic
1296 Material Formed by Methylglyoxal in Aqueous Aerosol Mimics. *Atmos. Chem. Phys.*
1297 10 (3), 997–1016.
- 1298 Schill, G. P., De Haan, M. A. Tolbert, 2014. Heterogeneous ice nucleation on simulated
1299 secondary organic aerosol, *Environ. Sci. Technol.* 48, 1675–1682.
- 1300 Sheesley, R. J., Schauer, J. J., Bean, E., Kenski, D., 2004. Trends in secondary organic aerosol at
1301 a remote site in Michigan's Upper Peninsula. *Environ. Sci. Technol.* 38, 6491–6500.
- 1302 Shen, R., Liu, Z., Liu, Y., Wang, L., Li, D., Wang, Y., Bai, Y., Li, X., 2018. Typical polar
1303 organic aerosol tracers in PM_{2.5} over the North China Plain: Spatial distribution,
1304 seasonal variations, contribution and sources. *Chemosphere*, 209, 758–766.
- 1305 Shiraiwa, M., Carslaw, N., Tobias, D.J., Waring, M.S., Rim, D., Morrison, G., Lakey, P.S.,
1306 Kruza, M., Von Domaros, M., Cummings, B.E. and Won, Y., 2019. Modelling
1307 consortium for chemistry of indoor environments (MOCCIE): integrating chemical

- 1308 processes from molecular to room scales. *Environ. Sci.: Process. Impacts* 21(8), 1240-
1309 1254.
- 1310 Shrivastava, M., Easter, R. C., Liu, X., Zelenyuk, A., Singh, B., Zhang, K., Ma, P.L., Chand, D.,
1311 Ghan, S., Jimenez, J. L., Zhang, Q., Fast, J., Rasch, P. J., Tiitta, P., 2015. Global
1312 transformation and fate of SOA: Implications of low-volatility SOA and gas-phase
1313 fragmentation reactions. *J. Geophys. Res.* 120, 4169–4195.
- 1314 Singh, P., Singh, M.K., Beg, Y.R., Nishad, G.R., 2019. A review on spectroscopic methods for
1315 determination of nitrite and nitrate in environmental samples. *Talanta* 191, 364-381.
- 1316 Sinha, V., Hakkim, H., Kumar, V., 2019. Advances in Identification and Quantification of Non-
1317 methane Volatile Organic Compounds Emitted from Biomass Fires through Laboratory
1318 Fire Experiments. *Atmos. Chem. Phys.* 2, 1-35.
- 1319 Song, M., Zhang, C., Wu, H., Mu, Y., Ma, Z., Zhang, Y., Liu, J., Li, X., 2019. The influence of
1320 OH concentration on SOA formation from isoprene photooxidation. *Sci. Total Environ.*
1321 650, 951-957.
- 1322 Sorooshian, A., Corral, A.F., Braun, R.A., Cairns, B., Crosbie, E., Ferrare, R., Hair, J., Kleb,
1323 M.M., Mardi, A.H., Maring, H., McComiskey, A., 2020. Atmospheric Research Over the
1324 Western North Atlantic Ocean Region and North American East Coast: A Review of Past
1325 Work and Challenges Ahead. *J. Geophys. Res-Atmos.* 125, e2019JD031626.
- 1326 Spichtinger, P., Gierens, K., 2009, Modelling of cirrus clouds—Part 1b: Structuring cirrus clouds
1327 by dynamics, *Atmos. Chem. Phys.* 9(2), 707–719.
- 1328 Spracklen, D.V., Jimenez, J.L., Carslaw, K.S., Worsnop, D.R., Evans, M.J., Mann, G.W., Zhang,
1329 Q., Canagaratna, M.R., Allan, J., Coe, H., McFiggans G., 2011. Aerosol mass
1330 spectrometer constraint on the global secondary organic aerosol budget. *Atmos. Chem.*
1331 *Phys.* 11 (23), 109-136.

- 1332 Sun, Y., Wang, Z., Fu, P., Jiang, Q., Yang, T., Li, J., Ge, X., 2013. The impact of relative
1333 humidity on aerosol composition and evolution processes during wintertime in Beijing,
1334 China. *Atmos. Environ.* 77, 927-934.
- 1335 Surratt, J. D., Murphy, S. M., Kroll, J. H., Ng, N. L., Hildebrandt, L., Sorooshian, A.,
1336 Szmigielski, R., Vermeylen, R., Maenhaut, W., Claeys, M., Flagan, R. C., Seinfeld, J.
1337 H., 2006. Chemical composition of secondary organic aerosol formed from the
1338 photooxidation of isoprene. *J. Phys. Chem. A* 110, 9665–9690.
- 1339 Surratt, J.D., Kroll, J.H., Kleindienst, T.E., Edney, E.O., Claeys, M., Sorooshian, A., Ng, N.L.,
1340 Offenberg, J.H., Lewandowski, M., Jaoui, M., Flagan, R.C., 2007. Evidence for
1341 organosulfates in secondary organic aerosol. *Environ. Sci. Technol.* 41(2), 517-527.
- 1342 Szmigielski, R., Surratt, J. D., Vermeylen, R., Szmigielska, K., Kroll, J. H., Ng, N. L., Murphy,
1343 S. M., Sorooshian, A., Seinfeld, J. H., Claeys, M., 2007. Characterization of 2-
1344 methylglyceric acid oligomers in secondary organic aerosol from the photooxidation of
1345 isoprene using trimethylsilylation and gas chromatography/ ion trap mass spectrometry.
1346 *J. Mass Spectrom.* 42, 101– 116.
- 1347 Tajuelo, M., Rodríguez, D., Baeza-Romero, M.T., Díaz-de-Mera, Y., Aranda, A., Rodríguez, A.,
1348 2019. Secondary organic aerosol formation from styrene photolysis and photooxidation
1349 with hydroxyl radicals. *Chemosphere* 231, 276-286.
- 1350 Takahama, S., Dillner, A.M., Weakley, A.T., Reggente, M., Bürki, C., Lbadaoui-Darvas, M.,
1351 Debus, B., Kuzmiakova, A., Wexler, A.S., 2019. Atmospheric particulate matter
1352 characterization by Fourier transform infrared spectroscopy: a review of statistical
1353 calibration strategies for carbonaceous aerosol quantification in US measurement
1354 networks. *Atmos. Meas. Tech. Discuss.* 12 (1), 525-567.
- 1355 Thepnuan, D., Chantara, S., Lee, C.T., Lin, N.H., Tsai, Y.I., 2019. Molecular markers for
1356 biomass burning associated with the characterization of PM_{2.5} and component sources

- 1357 during dry season haze episodes in Upper South East Asia. *Sci. Total Environ.* 658, 708-
1358 722.
- 1359 Tsay, S.C., Maring, H.B., Lin, N.H., Buntoung, S., Chantara, S., Chuang, H.C., Gabriel, P.M.,
1360 Goodloe, C.S., Holben, B.N., Hsiao, T.C., Hsu, N.C., 2016. Satellite-surface perspectives
1361 of air quality and aerosol-cloud effects on the environment: An overview of 7-
1362 SEAS/BASELInE. *Aerosol Air Qual. Res.* 16(11), 2581-2602.
- 1363 Tsigaridis, K., Kanakidou, M., 2003. Global modeling of secondary organic aerosol in the
1364 troposphere: a sensitivity analysis. *Atmos. Chem. Phys.* 3, 1849-1869.
- 1365 Updyke, K. M., Nguyen, T. B., Nizkorodov, S. A., 2012. Formation of brown carbon via
1366 reactions of ammonia with secondary organic aerosols from biogenic and anthropogenic
1367 precursors. *Atmos. Environ.* 63, 22–31.
- 1368 Vakkari, V., Beukes, J.P., Dal Maso, M., Aurela, M., Josipovic, M., Van Zyl, P.G., 2018. Major
1369 secondary aerosol formation in southern African open biomass burning plumes. *Nat.*
1370 *Geosci.*, 11(8), 580-583.
- 1371 Vidovic, K., Kroflič, A., Šala, M., Grgić, I., 2020. Aqueous-Phase Brown Carbon Formation
1372 from Aromatic Precursors under Sunlight Conditions. *Atmosphere*, 11(2), 131.
- 1373 Volkamer, R., Barnes, I., Platt, U., Molina, L.T., Molina, M.J., 2006. Remote sensing of glyoxal
1374 by differential optical absorption spectroscopy (DOAS): Advancements in simulation
1375 chamber and field experiments." *Environmental Simulation Chambers: Application to*
1376 *Atmospheric Chemical Processes.* Springer, Dordrecht, 129-141.
- 1377 Wagner, R., Möhler, O., Saathoff, H., Schnaiter, M., Skrotzki, J., Leisner, T., Wilson, T. W.,
1378 Malkin, T. L., Murray, B. J., 2012. Ice cloud processing of ultra-viscous/glassy aerosol
1379 particles leads to enhanced ice nucleation ability, *Atmos. Chem. Phys.*, 12, 8589–8610.

- 1380 Walhout, E. Q., Yu, H., Thrasher, C., Shusterman, J. M., O'Brien, R. E., 2019a. Effects of
1381 Photolysis on the Chemical and Optical Properties of Secondary Organic Material over
1382 Extended Time Scales. *ACS Earth Space Chem.* 3 (7), 1226–1236.
- 1383 Walhout, E.Q., Dorn, S.E., Martens, J., Berden, G., Oomens, J., Cheong, P.H.Y., Kroll, J.H.,
1384 O'Brien, R.E., 2019b. Infrared Ion Spectroscopy of Environmental Organic Mixtures:
1385 Probing the Composition of α -pinene Secondary Organic Aerosol. *Environ. Sci.*
1386 *Technol.* 53 (13), 7604-7612.
- 1387 Wallington, T.J., Seinfeld, J.H., Barker, J.R., 2019. 100 Years of progress in gas-phase
1388 atmospheric chemistry research. *Meteorol. Monogr.* 59, 10.1-10.59.
- 1389 Wang, C., Lei, Y. D., Endo, S., Wania, F., 2014. Measuring and modeling the salting-out effect
1390 in ammonium sulfate solutions. *Environ. Sci. Technol.* 48 (22), 238-245.
- 1391 Wang, S., Du, L., Tsona, N.T., Jiang, X., You, B., Xu, L., Yang, Z., Wang, W., 2020b. Effect of
1392 NO_x and SO₂ on the photooxidation of methylglyoxal: Implications in secondary aerosol
1393 formation. *J. Environ. Sci.* 92, 151-162.
- 1394 Wang, W., Kourtchev, I., Graham, B., Cafmeyer, J., Maenhaut, W., Claeys, M., 2005.
1395 Characterization of oxygenated derivatives of isoprene related to 2-methyltetrols in
1396 Amazonian aerosols using trimethylsilylation and gas chromatography/ion trap mass
1397 spectrometry. *Rapid Commun. Mass Spectrom.* 19(10), 1343-1351.
- 1398 Wang, W., Vas, G., Dommisse, R., Loones, K., Claeys, M., 2004. Fragmentation study of
1399 diastereoisomeric 2-methyltetrols, oxidation products of isoprene, as their trimethylsilyl
1400 ethers, using gas chromatography/ion trap mass spectrometry. *Rapid Commun. Mass*
1401 *Sp.* 18, 1787–179.
- 1402 Wang, X., C. L., Heald, A. J., Sedlacek, S. S., de Sá, S. T., Martin, M. L., Alexander, T. B.,
1403 Watson, A. C., Aiken, S., Springston, R., Artaxo, P., 2016. Deriving brown carbon
1404 from multiwavelength absorption measurements: Method and application to AERONET
1405 and Aethalometer observations. *Atmos. Chem. Phys.* 16 (19), 1733–752.

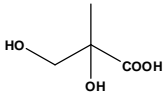
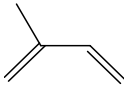
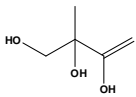
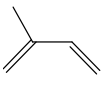
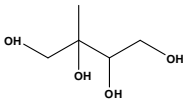
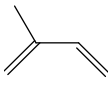
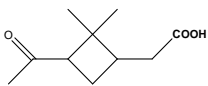



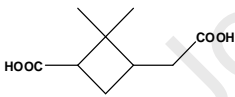

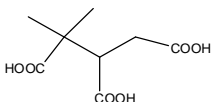
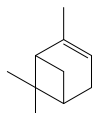
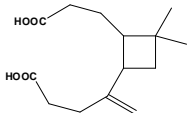
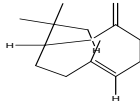
- 1406 Wang, Y., Chen, Y., Wu, Z., Shang, D., Bian, Y., Du, Z., Schmitt, S.H., Su, R., Gkatzelis, G.I.,
 1407 Schlag, P. and Hohaus, T., 2020a. Mutual promotion between aerosol particle liquid
 1408 water and particulate nitrate enhancement leads to severe nitrate-dominated particulate
 1409 matter pollution and low visibility. *Atmos. Chem. Phys.* 20(4), 2161-2175.
- 1410 Wang, Y., Pavuluri, C.M., Fu, P., Li, P., Dong, Z., Xu, Z., Ren, H., Fan, Y., Li, L., Zhang, Y.L.,
 1411 Liu, C.Q., 2019. Characterization of Secondary Organic Aerosol Tracers over Tianjin,
 1412 North China during Summer to Autumn. *ACS Earth Space Chem.* 3(10), 2339-2352.
- 1413 Washenfelder, R. A., Attwood, A. R., Brock, C. A., Hu, W., Baumann, K., Campuzano-Jost, P.,
 1414 Day, D. A., Edgerton, E. S., Murphy, D. M., Palm, B. B., McComiskey, A., Wagner, N.
 1415 L., de Sá, S. S., Ortega, A., Martin, S. T., Jimenez, J. L., Brown, S. S., 2015. Biomass
 1416 burning dominates brown carbon absorption in the rural southeastern United States.
 1417 *Geophys. Res. Lett.* 42, 653–664.
- 1418 Wennberg, P.O., Bates, K.H., Crounse, J.D., Dodson, L.G., McVay, R.C., Mertens, L.A.,
 1419 Nguyen, T.B., Praske, E., Schwantes, R.H., Smarte, M.D., StClair, J.M., Teng, A.P.,
 1420 Zhang, X., Seinfeld, J.H., 2018. Gas -phase reactions of isoprene and its major oxidation
 1421 products. *Chem. Rev.* 118 (7), 3337 -3390.
- 1422 Williams, B. J., Goldstein, A. H., Millet, D. B., Holzinger, R., Kreisberg, N. M., Hering, S. V.,
 1423 Allan, J. D., Worsnop, D. R., Jimenez, J. L., White, A. B., 2007. Chemical speciation of
 1424 organic aerosol during ICARTT 2004: results from in-situ measurements. *J. Geophys.*
 1425 *Res.* 112, 1-14.
- 1426 Wilson, T.W., Murray, B.J., Wagner, R., Mööhler, O., Saathoff, H., Schnaiter, M., Skrotzki, J.,
 1427 Price, H.C., Malkin, T.L., Dobbie, S., Al-Jumur, S.M.R.K., 2012. Glassy aerosols with a
 1428 range of compositions nucleate ice heterogeneously at cirrus temperatures, *Atmos. Chem.*
 1429 *Phys.* 12, 8611–8632.

- 1430 Winterhalter, R., Dingenen, R.V., Larsen, B.R., Jensen, N.R., Hjorth, J., 2003. LC-MS analysis
1431 of aerosol particles from the oxidation of α -pinene by ozone and OH-radicals. Atmos.
1432 Chem. Phys. Discuss. 3(1), 1-39.
- 1433 Wong, J.P., Tsagkaraki, M., Tsiodra, I., Mihalopoulos, N., Violaki, K., Kanakidou, M., Sciare,
1434 J., Nenes, A., Weber, R. J., 2019. Atmospheric evolution of molecular-weight-separated
1435 brown carbon from biomass burning. Atmos. Phys. 19 (11), 7319-7334.
- 1436 Wu, Z., Hu, L., Guo, T., Lin, T., Guo, Z., 2020. Aeolian transport and deposition of
1437 carbonaceous aerosols over the Northwest Pacific Ocean in spring. Atmos. Environ. 223,
1438 117209.
- 1439 Xia, M., Wang, W., Wang, Z., Gao, J., Li, H., Liang, Y., Yu C., Zhang, Y. Wang P, Zhang Y, Bi
1440 F., 2019. Heterogeneous Uptake of N_2O_5 in Sand Dust and Urban Aerosols Observed
1441 during the Dry Season in Beijing. Atmosphere, 10 (4), 1-16.
- 1442 Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-
1443 VanWertz, G., Kreisberg, N. M., Knote, C., Olson, K., Koss, A., Goldstein, A. H.,
1444 Hering, S. V., de Gouw, J., Baumann, K., Lee, S., Nenes, A., Weber, R. J., Ng, N. L.,
1445 2015. Effects of anthropogenic emissions on aerosol formation from isoprene and
1446 monoterpenes in the southeastern United States. P Natl. Acad. Sci. USA, 112, 37–42.
- 1447 Yu, J., Griffin, R. J., Cocker III, D. R., Flagan, R. C., Seinfeld, J. H., Blanchard, P., 1999.
1448 Observation of gaseous and particulate products of monoterpene oxidation in forest
1449 atmospheres. Geophys. Res. Lett. 26, 1145–1148.
- 1450 Yu, X., Shen, L., Hou, X., Yuan, L., Pan, Y., An, J. and Yan, S., 2020. High-resolution
1451 anthropogenic ammonia emission inventory for the Yangtze River Delta,
1452 China. Chemosphere 251, 126342.
- 1453 Yuan, J., Ling, Z., Wang, Z., Lu, X., Fan, S., He, Z., Guo, H., Wang, X., Wang, N., 2018. PAN–
1454 Precursor Relationship and Process Analysis of PAN Variations in the Pearl River Delta
1455 Region. Atmosphere, 9(10), 372.

- 1456 Zare, A., Fahey, K. M., Sarwar, G., Cohen, R. C., Pye, H. O. T., 2019. Vapor-Pressure Pathways
1457 Initiate but Hydrolysis Products Dominate the Aerosol Estimated from Organic Nitrates.
1458 ACS Earth Space Chem. 3 (8), 1426–1437.
- 1459 Zaveri, R.A., Shilling, J.E., Fast, J.D., Springston, S.R., 2020. Efficient Nighttime Biogenic SOA
1460 Formation in a Polluted Residual Layer. J. Geophys. Res-Atmos. 125(6), 2019JD031583.
- 1461 Zhang, F., Yu, X., Chen, J., Zhu, Z. and Yu, X.Y., 2019a. Dark air–liquid interfacial chemistry
1462 of glyoxal and hydrogen peroxide. NPJ Clim. Atmos. Sci. 2(1), 1-8.
- 1463 Zhang, Q., Jimenez, J., Canagaratna, M., Ulbrich, I., Ng, N., Worsnop, D., Sun, Y., 2011.
1464 Understanding atmospheric organic aerosols via factor analysis of aerosol mass
1465 spectrometry: a review. Anal. Bioanal. Chem. 401 (10), 3045–3067.
- 1466 Zhang, Q., Song, Y., Li, M., Zheng, B., 2020a. Anthropogenic Emissions of SO₂, NO_x, and NH₃
1467 in China. In Atmospheric Reactive Nitrogen in China (pp. 13-40). Springer, Singapore.
1468 https://doi.org/10.1007/978-981-13-8514-8_2.
- 1469 Zhang, Q.I., Jimenez, J.L., Worsnop, D.R., Canagaratna, M., 2007. A case study of urban
1470 particle acidity and its influence on secondary organic aerosol. Environ. Sci. Technol.
1471 41, 3213e3219.
- 1472 Zhang, T., Yang, W., Han, C., Yang, H., Xue, X., 2020b. Heterogeneous reaction of ozone with
1473 syringic acid: Uptake of O₃ and changes in the composition and optical property of
1474 syringic acid. Environ. Pollut. 257, 113632.
- 1475 Zhang, X., Kim, H., Parworth, C. L., Young, D. E., Zhang, Q., Metcalf, A. R., Cappa, C. D.,
1476 2016. Optical properties of wintertime aerosols from residential wood burning in

- 1477 Fresno, CA: Results from DISCOVER-AQ 2013, *Environ. Sci. Technol.* 50 (4), 1681–
1478 1690.
- 1479 Zhang, Y., Chen, Y., Lei, Z., Olson, N. E., Riva, M., Koss, A. R., Zhang, Z., Gold, A., Jayne, J.
1480 T., Worsnop, D. R., Onasch, T. B., Kroll, J. H., Turpin, B. J., Ault, A. P., Surratt, J. D.,
1481 2019b. Joint Impacts of Acidity and Viscosity on the Formation of Secondary Organic
1482 Aerosol from Isoprene Epoxydiols (IEPOX) in Phase Separated Particles. *ACS Earth*
1483 *Space Chem.* 3 (12), 2646–2658.
- 1484 Zhou, W., Mekic, M., Liu, J., Loisel, G., Jin, B., Vione, D., Gligorovski, S., 2019. Ionic strength
1485 effects on the photochemical degradation of acetosyringone in atmospheric deliquescent
1486 aerosol particles. *Atmos. Environ.* 198, 83-88.
- 1487 Zhu, J., Penner, J.E., Yu, F., Sillman, S., Andreae, M.O., Coe, H., 2019. Decrease in radiative
1488 forcing by organic aerosol nucleation, climate, and land use change. *Nat. Commun.* 10
1489 (1), 423, 1-7.
- 1490 Zhu, R.G., Xiao, H.Y., Lv, Z., Xiao, H., Zhang, Z. and Xiao, H., 2020. Nitrogen isotopic
1491 composition of free Gly in aerosols at a forest site. *Atmos. Environ.* 222, 117179.
- 1492 Zhu, W., Luo, L., Cheng, Z., Yan, N., Lou, S., Ma, Y., 2018. Characteristics and contributions of
1493 biogenic secondary organic aerosol tracers to PM_{2.5} in Shanghai, China. *Atmos. Pollut.*
1494 *Res.* 9(2),179-188.
- 1495 Zobrist, B., Marcolli, C., Peter, T., Koop, T., 2008. Heterogeneous ice nucleation in aqueous
1496 solutions: The role of water activity, *J. Phys. Chem. A*, 112, 3965–3975.

Table 1. BSOA, Precursors, mass fragments and their day & night-time dominance

S.N.	SOA Tracer Name	Precursor	Mass of SOA	Mass Fragments	Mechanism of formation Dominated in	References
1.	 2-Methylglyceric acid	 Isoprene	134	321, 203, 293, 219, 337	Day	Fu et al., 2010
2.	 C5-Alkene triols	 Isoprene	118	NA	Day	Wang et al., 2005; Kourtchev et al., 2005
3.	 2-Methyltetrols	 Isoprene	180	409, 219, 319, 293, 203	Day	Edney et al., 2005; Claeys et al., 2004,
4.	 <i>cis</i> -Pinonic acid	 Terpene	184	257, 121, 139, 167, 187	Day	Fu et al., 2010
5.	 <i>cis</i> -Norpinic acid	 Terpene	NA	NA	Night	Fu et al., 2010
6.	 Pinic acid	 Terpene	186	241, 315, 151, 197, 331	Night	Fu et al., 2010
7.	 MBTCA ^a	 Terpene	NA	NA	Night	Fu et al., 2010
8.	 β -Caryophyllinic acid	 β -caryophylline	254	309, 383, 399, 427, 439	Day & Night	Fu et al., 2010

^a3-methyl-1,2,3-butanetricarboxylic acid

Table 2. Concentration (ng m⁻³) of BSOA form difference studies

S.N	Study site	Event	Particle Size	Isoprene SOA tracers				Monoterpenes SOA tracers			β -caryophyllin	References
				MGA	C5-alkene triols	MTL	3-HGA	PA	Pinic acid	MBTCA	CPA	
1.	Qinghai Lake	Summer	TSP	1.0±0.35	1.2±0.81	1.6±0.84	3.7±2.1	11±4.2	0.72±0.23	0.22±0.22	ND	Ren et al., 2018
2.	Shanghai, China	Winter	TSP	2.5±0.58	0.44±0.24	0.81±0.29	4.4±1.2	8.0±1.7	3.9±1.2	ND	ND	Ren et al., 2018
3.	Alaska, USA	Summer	TSP	0.5±0.1	7.7±2.1	15.2±3.1	0.59±0.07	1.34±0.6	5.18±1.2	2.65±0.3	0.62±0.1	Haque et al., 2016
4.	Alaska, USA	Winter	TSP	0.09	0.02	0.04	0.23	0.68	1.3	0.1	0.04	Haque et al., 2016
5.	Mumbai, India	Winter	PM ₁₀	1.2±0.80	0.95±1.2	1.4±0.45	14±11	1.9±0.73	2.7±1.9	0.49±0.37	5.6±2.9	Fu et al., 2016
6.	Chennai, India	Winter	PM ₁₀	0.67±0.45	2.18±2.93	2.75±2.78	6.24±6.68	0.82±0.36	1.00±0.82	0.25±0.30	2.76±1.92	Fu et al., 2010
7.	Central China	Annual	PM ₁₀	4.1±2.2	8.1±9.8	8.5±6.4	2.1±1.1	3.0±1.4	NA	1.6±1.1	2.2±1.2	Li et al., 2013
8.	South China	Fall-winter	PM _{2.5}	2.04±1.9	NA	27.6±15.1	0.43±0.59	3.60±3.76	1.25±0.79	1.16±0.99	0.54±0.56	Ding et al., 2011
9.	Beijing, China	Winter	PM _{2.5}	3.63	2.38	1.17	4.27	NA	NA	NA	NA	Shen et al., 2018
10.	Tianjin, China	Summer	PM _{2.5}	20.09	15.32	34.42	15.58	NA	NA	NA	NA	Shen et al., 2018
11.	Shanghai, China	Summer-Spring	PM _{2.5}	1.29±0.92	NA	6.07±3.86	0.39±0.46	0.92±0.93	NA	1.45±1.17	0.58±0.75	Zhu et al., 2018
12.	Guanzhou	Annual	PM _{2.5}	2.05±2.07	17.7±22.2	8.14±10.8	11.8±8.09	5.82±5.92	1.34±0.92	9.25±7.94	9.25±7.94	Yuan et al., 2018
13.	East China	Summer	TSP	1.4	2.2	4.8	2.2	NA	3.4	5.6	2.9	Kang et al., 2018
14.	Jinan, China	Annual	PM _{2.5}	7.5±12.6	8.8±24.2	26.7±56.6	11.6±6.2	12.4±5.0	NA	NA	NA	Liu et al., 2019a
15.	Southeastern China	Annual	PM _{2.5}	1.38±0.72	11.3±18.1	32.5±48.8	13.3±9.36	0.41±0.95	1.22±1.25	7.8±6.08	5.99±7.25	Hong et al., 2019

NA – Not Available, ND- Not Detected

Highlights

- The BSOA exhibit distinct day and nighttime formation mechanism.
- Analytical advancements and their ability to examine SOA are compiled and compared.
- SOA can cause significantly enhanced radiating forcing.
- Anthropogenic gaseous pollutants have strong influence over BSOA formation.
- Meteorological variables can effectively control BSOA formation.

Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Figure Captions

Fig.1. BSOA formation pathways via biogenic volatile organic compounds.

Fig.2. Atmospheric degradation pathway of biogenic volatile organic compounds.

Fig.3. Gas phase isoprene oxidation and formation of first and second-generation products. Here red box indicates night time dominated process and green box indicated day time dominated process

Fig.4. 3D depiction of some of the current techniques used to analyse the aerosol's organic content on basis of their supplemental nature. The explanation and abbreviations are given in the text.

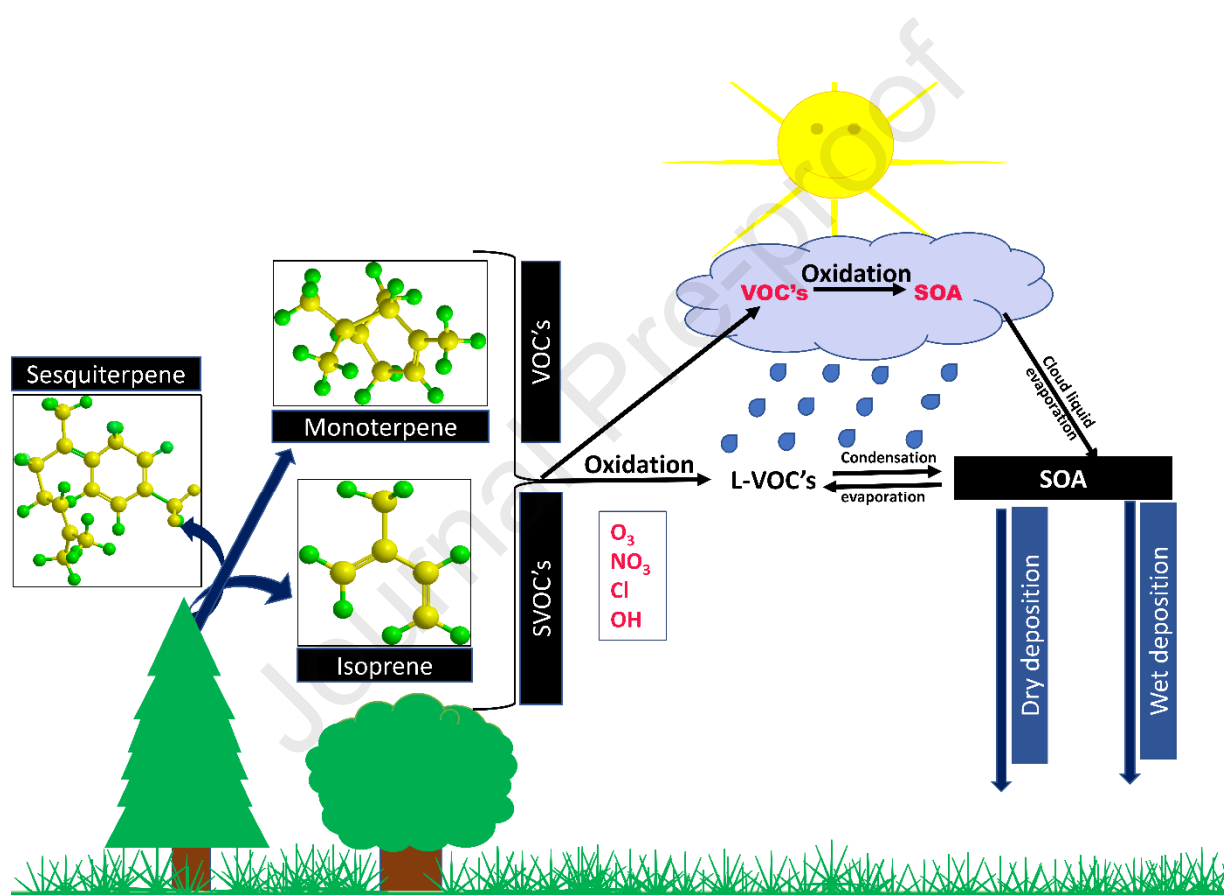


Fig.1.

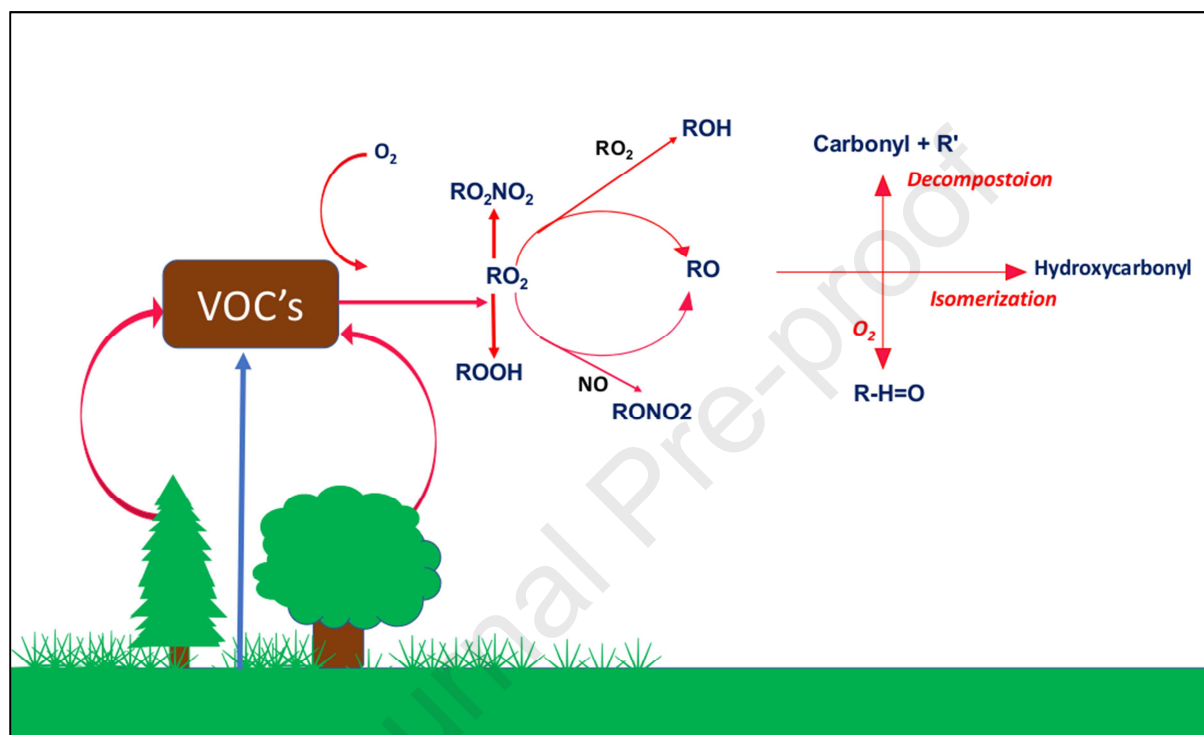


Fig.2.

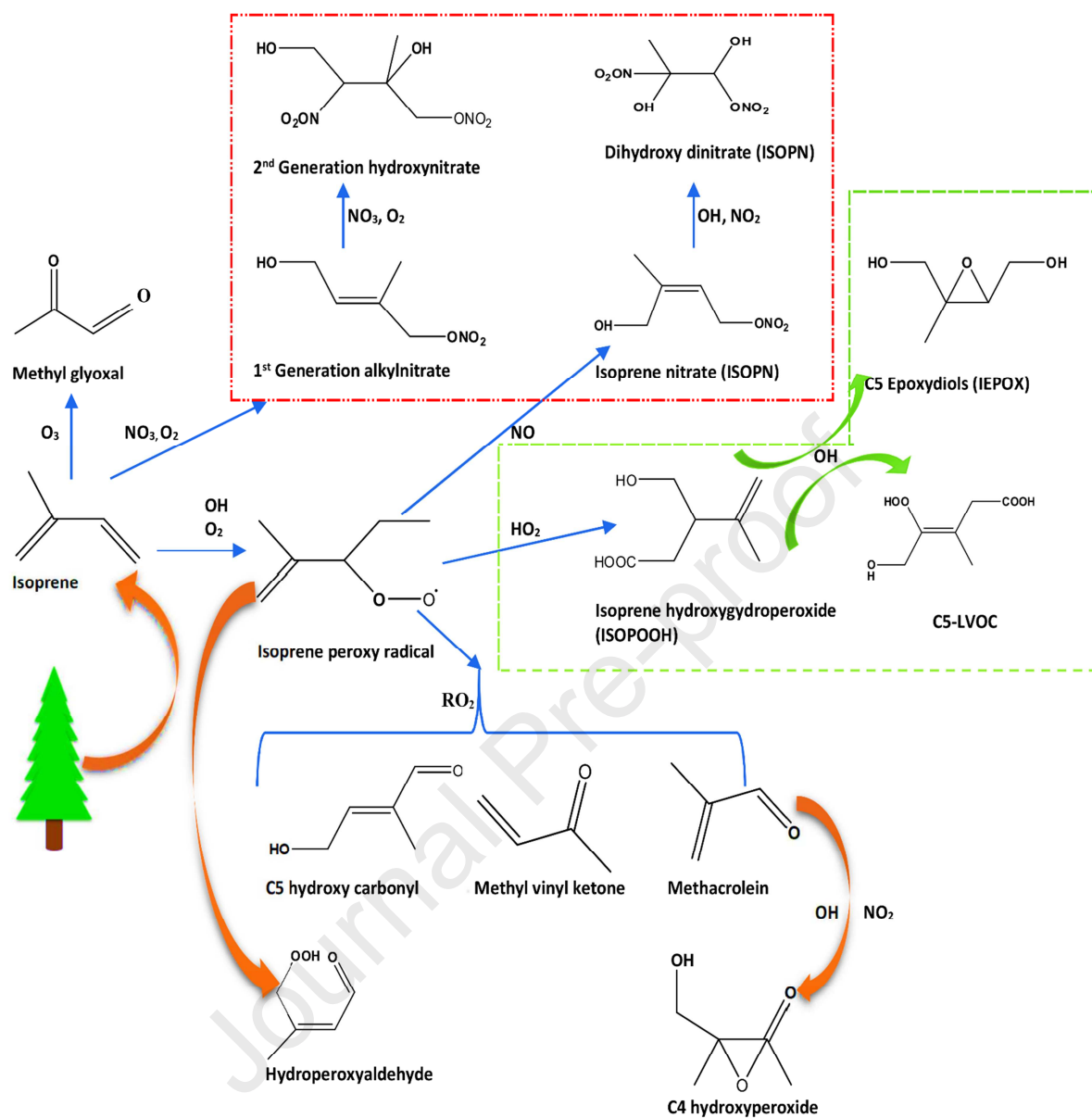


Fig.3.

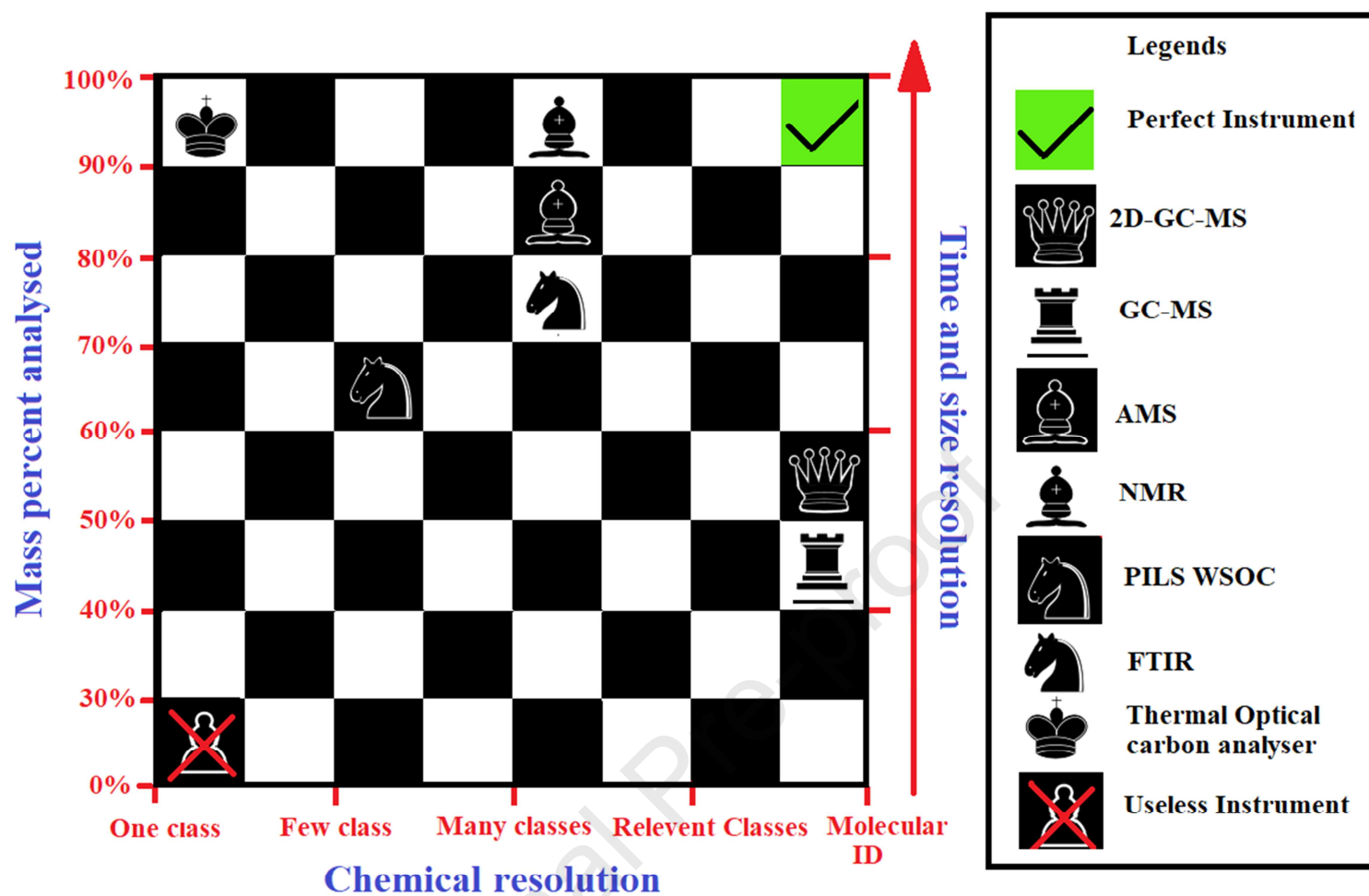


Fig.4.