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Key Points:
- Recent progresses include development of instruments and observations of multicomponent nucleation and NPF at various locations
- There is a lack of understanding of fundamental chemical mechanisms responsible for NPF in diverse environments, especially in megacities
- There is a need to develop NPF parameterizations to represent a wider range of pollutants, temperature and humidity

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New Particle Formation in the Atmosphere: From Molecular Clusters to Global Climate

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Abstract New particle formation (NPF) represents the first step in the complex processes leading to formation of cloud condensation nuclei. Newly formed nanoparticles affect human health, air quality, weather, and climate. This review provides a brief history, synthesizes recent significant progresses, and outlines the challenges and future directions for research relevant to NPF. New developments include the emergence of state-of-the-art instruments that measure prenucleation clusters and newly nucleated nanoparticles down to about 1 nm; systematic laboratory studies of multicomponent nucleation systems, including collaborative experiments conducted in the Cosmics Leaving Outdoor Droplets chamber at CERN; observations of NPF in different types of forests, extremely polluted urban locations, coastal sites, polar regions, and high-elevation sites; and improved nucleation theories and parameterizations to account for NPF in atmospheric models. The challenges include the lack of understanding of the fundamental chemical mechanisms responsible for aerosol nucleation and growth under diverse environments, the effects of SO_2 and NO_x on NPF, and the contribution of anthropogenic organic compounds to NPF. It is also critical to develop instruments that can detect chemical composition of particles from 3 to 20 nm and improve parameterizations to represent NPF over a wide range of atmospheric conditions of chemical precursor, temperature, and humidity.

Plain Language Summary In the atmosphere, invisible to the human eye, there are many microscopic particles, or "nanoparticles," that affect human health, air quality, and climate. We do not fully understand the chemical processes that allow these fine particles to form and be suspended in the air nor how they influence heat flow in Earth's atmosphere. Laboratory experiments, field observations, and modeling simulations have all shown different results for how these particles behave. These inconsistencies make it difficult to accurately represent the processes of new particle formation in regional and global atmospheric models. Scientists still need to develop instruments that can measure the smallest range of nanoparticles and to find ways to describe particle formation that allow for differences in temperature, humidity, and level of pollution.

1. Introduction

Atmospheric aerosols, which are defined as liquid or solid particles suspended in air, have profound impacts on the Earth-atmosphere system. The aerosol effects include air quality, human health, ecosystem, weather, and climate, all of which have important societal implications (IPCC, 2013; NASEM, 2016). For example, aerosol particles absorb and scatter incoming solar radiation in the atmosphere, leading to important consequences for the Earth's energy budget. This effect, commonly referred to as aerosol-radiation interaction, contributes importantly to cooling (by scattering) or warming (by absorption) in the atmosphere (IPCC, 2013; Peng et al., 2016; Y. Wang et al., 2013). By acting as cloud condensation nuclei (CCN) and ice nuclei, aerosols also play an important role in controlling cloud formation/development, precipitation efficiency, and the albedo, frequency, and lifetime of clouds (Albrecht et al., 1989; Fan et al., 2018; Li et al., 2008; Rosenfeld et al., 2008; Seinfeld & Pandis, 2016; Twomey, 1974). These effects are often referred to collectively as aerosol-cloud interactions, which are also critical to radiative transfer in the atmosphere. Presently, the
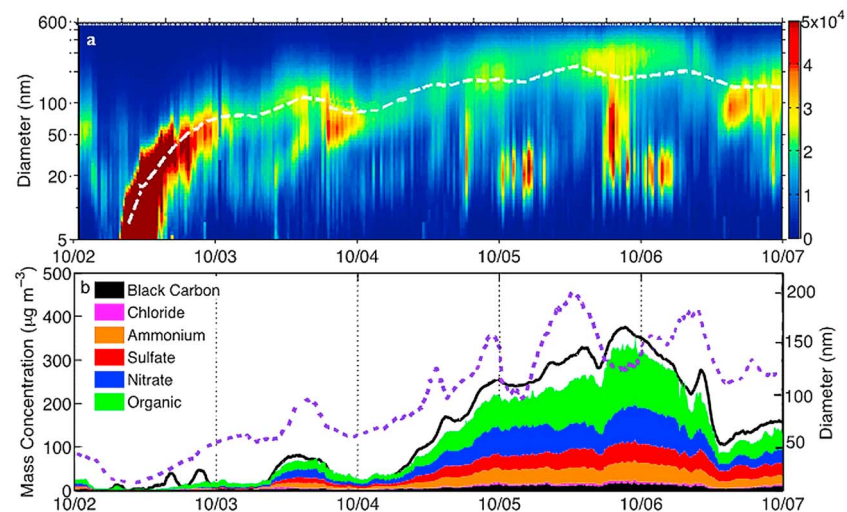
Figure 1. Aerosol nucleation and growth during a pollution episode (2–7 October, 2013) in Beijing. (a) Temporal evolutions of particle number size distribution and mean diameter (white dashed curve). (b) PM$_{2.5}$ mass concentration (black solid line), mean diameter (purple dashed line), and PM$_1$ (particulate matter smaller than 1.0 μm) chemical composition. The shaded colors denote the mass concentrations of the aerosol constituents, that is, green for organics, blue for nitrate, red for sulfate, yellow for ammonium, purple for chloride, and black for black carbon. Adapted from Guo et al. (2014).

aerosol effects associated with aerosol-radiation interaction and aerosol-cloud interaction represent the largest uncertainty in predictions of anthropogenic forcing on climate (IPCC, 2013). In addition, fine aerosols (i.e., particulate matter smaller than 2.5 μm or PM$_{2.5}$) constitute a key component of air pollution on local and regional scales, since high concentrations of PM$_{2.5}$ not only cause visibility degradation but also negatively impact human health (EPA, 2004). More than 3 million premature deaths each year worldwide have been attributed to exposure to fine PM, representing one of the greatest risks to public health (Lim et al., 2012). There is increasing evidence that exposure to fine aerosols not only causes acute and chronic diseases but also exacerbates adverse health effects of gaseous criteria pollutants, including ozone (O$_3$), sulfur dioxide (SO$_2$), carbon monoxide (CO), and nitrogen oxides (NO$_x$ = NO + NO$_2$; Kimmel et al., 1997; Künzli et al., 2005). In addition, it has been suggested that ultrafine particles likely exert the most severe health effects, since small particles are more likely to be deposited in the pulmonary region and to penetrate into bloodstream than large particles (Anderson et al., 2012; Araujo et al., 2008; Hamra et al., 2014; NASEM, 2016; Oberdorster et al., 2004; Schlesinger et al., 2006).

Aerosol particles are either emitted directly into the atmosphere (referred to as primary particles) or produced in the atmosphere via gas-to-particle conversion (referred to as secondary particles; Seinfeld & Pandis, 2016). In addition, primary and secondary aerosols undergo chemical and physical transformations and are subjected to transport, cloud processing, and removal from air (Zhang et al., 2015). New particle formation (NPF), which is characterized by a sudden burst of high concentrations of subnanometer-sized (1–3 nm) particles in the atmosphere followed by their growth (Figure 1), represents a major source for tropospheric aerosol population. NPF events have been observed under diverse environmental conditions, including urban locations, forested areas, marine/coastal regions, and the remote or free troposphere (Kerminen et al., 2018; Kulmala, Vehkamäki, et al., 2004; Z. Wang et al., 2017). Atmospheric observations show that about 10% to 60% of the NPF events lead to CCN formation and the average enhancement factors in CCN concentrations due to NPF range from 0.5 to 11 depending on the location (Asmi et al., 2011; Dameto de España et al., 2017; Kerminen et al., 2018; Kuang et al., 2009; Laakso et al., 2013; Lihavainen et al., 2003; Rose et al., 2017; Shen et al., 2016; Sihto et al., 2011; Z. B. Wang, Hu, Sun, et al., 2013; Yu et al., 2014; Yue et al., 2011). Global modeling predictions also show that NPF contributes around half of the CCN population in the troposphere (Gordon et al., 2017; Merikanto et al., 2009; Yu et al., 2014; Yu & Luo, 2009; Yue et al., 2011). NPF has been shown to be also associated with severe haze events in Chinese megacities (Figure 1), producing a large number of nanoparticles that subsequently grow into submicron-sized particles (e.g., CCN size) even under polluted environments (Guo et al., 2014).

NPF occurs in two stages (Kulmala et al., 2013; Zhang, 2010). The first step involves the formation of a critical nucleus during the phase transformation from vapor to liquid or solid. The second step is the growth of the critical nucleus to a larger size (>2–3 nm) that competes with removal by preexisting aerosols. The nucleation process results in a decrease in both enthalpy and entropy (i.e., $\Delta H < 0$ and $\Delta S < 0$). A free-energy barrier (i.e., $\Delta G = \Delta H - T\Delta S > 0$) needs to be overcome before the phase transition becomes spontaneous. An additional limitation in the growth of nanoparticles is related to highly elevated vapor pressures over small clusters and nanoparticles. This phenomenon is commonly known as the Kelvin or curvature effect, which imposes another major restriction in the growth of freshly nucleated nanoparticles.

The mechanisms for nucleation have been widely investigated in many interdisciplinary areas, including biomedicine, catalysis, fuel cell, magnetic data storage, agriculture, and solar cells, because of the broad applications of nanoparticles (Thanh et al., 2014). These nanoparticles have unique magnetic, optical, electronic, and catalytic properties, which are size-dependent and distinct from the bulk materials. Nucleation is generally defined as the creation of molecular clusters prior to spontaneous formation of a new phase from vapor, liquid, to solid transition (Lamer & Dinegar, 1950). Formation of the new phase without heterogeneities occurs through random fluctuations in vapor density, generating molecular clusters that grow or decay by gaining or losing a monomer. Cluster growth occurs via a reversible, stepwise kinetic process for single or multicomponent systems. Several theoretical frameworks have been developed to describe the rates and mechanism by which clusters grow or decay. The phenomenological classical nucleation theory (CNT) considers the thermodynamics and kinetics of nucleation, by evaluating the change in Gibbs free energy (ΔG) for the formation a cluster of radius r (Flood, 1934; Reiss, 1950),

$$\Delta G = -nkTlnS' + 4\pi r^2 \sigma, \tag{1}$$

where $S' = p/p_S$ is the saturation ratio, p is the vapor pressure, p_S is the saturation pressure over a plane surface, and σ is the surface tension. The CNT utilizes the capillarity approximation, that is, assuming that the cluster possesses identical particle density, surface tension, and equilibrium vapor pressure as the bulk. For a spherical cluster, the number of molecules n is related to its radius $n = (4/3)\pi r^3 / v_l$, where v_l is the volume of a single molecule. The free energy reaches a maximum at the critical nucleus

$$\Delta G^* = \frac{4\pi}{3}\sigma r^{*2} = \frac{16\pi}{3}\frac{\sigma^3 v_l^2}{\left(kT \ln S'\right)^2}. \tag{2}$$

The ΔG^* also corresponds to the minimum free energy required to form stable clusters. The critical radius (r^*) is defined as the minimum size at which a cluster survives evaporation

$$r^* = \frac{2\sigma v_l}{kT \ln S'}. \tag{3}$$

The critical nucleus is in a metastable state between the vapor and liquid, since addition of a monomer leads to spontaneous growth, while removal of a monomer results in decomposition of the critical nucleus. The nucleation rate (J), which is defined as the number of stable clusters formed per unit volume per unit time (N), is expressed by an Arrhenius type equation,

$$J = \frac{dN}{dt} = J_o \exp\left(-\frac{\Delta G^*}{kT}\right), \tag{4}$$

where J_o is the preexponential factor relevant to gaseous kinetics. Major uncertainties of CNT lie in several simple assumptions, including that the critical nucleus has a spherical shape and its physical properties are identical to those of the bulk phase. However, many of the problems with CNT (Girshick & Chiu, 1990; Olenius & Riipinen, 2017; Wilemski & Wyslouzil, 1995; Yu, 2003) can be overcome, for example, with improved modeling of sulfuric acid hydrates (Noppel et al., 2001; Vehkamäki et al., 2002) or quantum chemical calculations (Du et al., 2009; McGrath et al., 2012).

The nucleation process can be formulated on the basis of the kinetics for growth and evaporation of clusters by a reversible, stepwise kinetic process, according to the following expression (Seinfeld & Pandis, 2016):
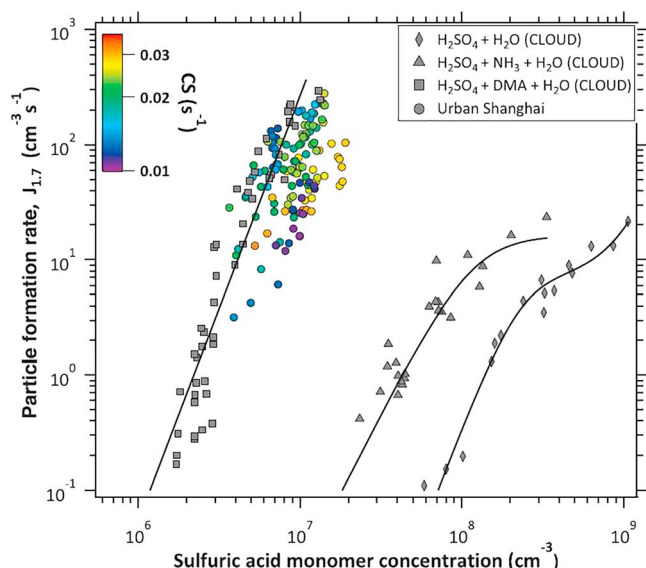
Figure 2. Measured particle formation rates for 1.7-nm particles ($J_{1.7}$) as a function of sulfuric acid concentrations and condensation sink (CS) in urban Shanghai, China (colored circles). In comparison, binary (diamonds) and ternary nucleation with ammonia (triangles; Kirkby et al., 2011) and diamine (squares; Almeida et al., 2013) experimental results taken from CLOUD chamber experiments are also included. Adapted from L. Yao et al. (2018).

$$\frac{d[C_i]}{dt} = k_{i-1}^+[C_{i-1}][A_{i-1}] - k_i^-[C_i] - k_i^+[C_i][A_i] + k_{i+1}^-[C_{i+1}], \qquad (5)$$

where A_{i-1} denotes a monomer species to be added to the cluster C_{i-1} at the $(i-1)$th step, k_i^- and k_i^+ represent the evaporation and growth rate constants of the cluster, respectively, and $[C_i]$ and $[A_i]$ denote the concentrations of the cluster and monomer at the step i, respectively. Under the steady-state condition, the cluster concentrations of different sizes become time-independent, since the net rate to form each cluster is constant. J is related to the growth (k_i^+) and evaporation (k_i^-) rate constants for each cluster, which are typically calculated from the gas-kinetic collision rate and the cluster stability (i.e., the free-energy change for cluster formation), respectively. Several kinetic approaches have been developed to calculate cluster distributions and J by obtaining the rate constants of formation and decomposition of clusters without explicit expressions of the formation energy and surface tension of clusters. In addition, the dynamical nucleation theory treats nucleation on the basis of multistep binary collisions between nucleating molecules and clusters (Schenter et al., 1999). Furthermore, microscopic schemes, including molecular dynamics and Monte Carlo calculations, have been employed to predict the structure and free energy of cluster formation according to the first principles (Seinfeld & Pandis, 2016; Zhang et al., 2012).

The nucleation theorem is a thermodynamic expression to correlate the nucleation barrier height to the change in log vapor concentration (Kashchiev, 1982). The kinetic formulation of the nucleation theorem, which can be derived by differentiating the steady-state J (equation (4)) at constant temperature, is readily generalized from a single component to a multicomponent system with a similar form (Oxtoby & Kashchiev, 1994)
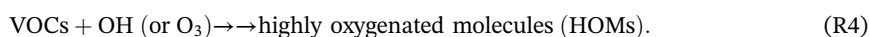
$$\frac{\partial \ln J_j}{\partial \ln S_j'} = \frac{\partial \ln J_j}{\partial \ln n_j} = g_j^* + \delta_j, \qquad (6)$$

where $\delta_j = J_j/J$ (i.e., $0 \le \delta_j \le 1$) and J_i, S_j', n_j, and g_j^* are the nucleation rate, the saturation ratio, the monomer number concentration, and the critical monomer number (in monomeric units), respectively, for species j. The kinetic nucleation theorem has been widely applied to infer molecular compositions of the critical nucleus on the basis of field and laboratory measured J and vapor concentrations (McGraw & Zhang, 2008; McMurry et al., 2005) but must be applied with care (Ehrhart & Curtius, 2013; Malila et al., 2015; Vehkamäki et al., 2012).

Extensive efforts have been made to elucidate the fundamental mechanisms relevant to atmospheric NPF on the basis of field measurements, laboratory experiments, and theoretical calculations (Kulmala et al., 2013; Zhang et al., 2012). Recent field studies include ambient measurements of ultrafine particles down to the size ~1 nm, gaseous concentrations of nucleating precursors (such as sulfuric acid H_2SO_4, ammonia NH_3, and amines), and prenucleation clusters (e.g.,Yu & Lee, 2012 ; Zhao et al., 2011 ; Zheng et al., 2015). For example, L. Yao et al. (2018) recently showed NPF in a very polluted Chinese megacity, Shanghai (Figure 2). They measured particle number size distributions down to ~1.2 nm, along with sulfuric acid vapor, sulfuric acid clusters (trimer and tetramer), and sulfuric acid-dimethylamine (DMA) clusters with up to four molecules of sulfuric acid and two molecules of DMA. Also, numerous laboratory experiments were conducted using a flow reactor to monitor nucleation at a fixed time or a reaction chamber to follow the evolution of nucleation (Yu, McGraw, & Lee, 2012; Zhang et al., 2009; Zhao et al., 2011). Notably, a major infrastructure, the Cosmics Leaving Outdoor Droplets (CLOUD) chamber, was established at CERN, Switzerland. The CLOUD experiment aims to understand aerosol nucleation and subsequent particle growth by integrating advanced instrumentation for measuring gases and aerosols with a high energy physics accelerator (proton synchrotron; Kirkby et al., 2011; Riccobono et al., 2014; Tröstl et al., 2016). In addition, theoretical investigations of aerosol nucleation were carried out to determine the stability and dynamics of prenucleation clusters

using thermodynamic data from quantum chemical calculations (Xu & Zhang, 2012), the Atmospheric Cluster Dynamics Code (ACDC) model (McGrath et al., 2012; Olenius et al., 2013; Zhao et al., 2009), and kinetic modeling calculations (Lovejoy et al., 2004; Yu, 2010, 2011).

NPF is intrinsically connected to gasphase chemistry, since many of the chemical species relevant to aerosol nucleation and growth are produced from photooxidation of atmospheric trace gases, including SO_2 and volatile organic compounds (VOCs),

$$SO_2 + OH \rightarrow HSO_3, \tag{R1}$$

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2, \tag{R2}$$

$$SO_3 + H_2O + M \rightarrow H_2SO_4 + M, \tag{R3}$$

$$VOCs + OH \ (or \ O_3) \rightarrow \rightarrow highly \ oxygenated \ molecules \ (HOMs). \tag{R4}$$

The critical role of photochemistry in NPF is also reflected by the fact that most NPF events occur during the day, while nocturnal NPF is also observed (Junninen et al., 2008). Sulfuric acid is considered the most prevalent nucleating species because of its low vapor pressure under typical atmospheric conditions (Sipila et al., 2010; Zhang et al., 2012). Also, sulfuric acid is prone to form hydrogen bonding with many atmospherically important compounds, including water, basic species (i.e., ammonia and amines), and organic acids. This ability to form hydrogen bonding renders sulfuric acid an ideal candidate to participate in nucleation. In particular, the prenucleation clusters exist in a state that is distinct from the liquid phase, and the very first step of nucleation (i.e., the formation of a dimer) is driven mainly by hydrogen bond. Field measurements showed that gaseous sulfuric acid in concentrations of greater than approximately 10^5 molecules/cm^3 is necessary for observable NPF in the atmosphere (Erupe et al., 2010; McMurry et al., 2005; Nieminen et al., 2009). In addition, several other nucleating precursors have been implicated for NPF under diverse environments, including ions (Kirkby et al., 2011; Kirkby et al., 2016), ammonia/amines (Almeida et al., 2013; Glasoe et al., 2015; Jen, Zhao, et al., 2016; Kirkby et al., 2011; Kürten et al., 2014; Yao et al., 2018; Yu & Luo, 2014a; Yu, McGraw, & Lee, 2012), HOMs (Ehn et al., 2014; Jokinen et al., 2015; Tröstl et al., 2016), other organic compounds (Metzger et al., 2010; Zhang et al., 2004), and iodine oxides (O'Dowd et al., 2002; Sipilä et al., 2016).

Currently, the mechanism and chemical species responsible for atmospheric NPF are still highly uncertain. In particular, consistent chemical mechanisms to explain NPF under diverse atmospheric conditions are still lacking. Also, available results of NPF from laboratory experiments, field measurements, and theoretical calculations are often conflicting, regarding the identity of the chemical species responsible for aerosol nucleation and growth under diverse ambient conditions. These inconsistencies are partially attributed to difficulties in detecting the nucleating precursor species and the chemical composition of clusters and freshly nucleated nanoparticles by analytical instrumentation and in calculating large prenucleation clusters by reliable theoretical methods.

The present review provides a synthetic view of NPF in the atmosphere, by emphasizing the most recent advances made in the understanding of nucleation and growth of atmospheric nanoparticles from laboratory experiments, field measurements, and modeling studies. The review is organized according to the following outline. Section 2 emphasizes on recent development of analytical instruments to measure the size and chemical composition of prenucleation clusters and nanoparticles. Recent progresses on atmospheric NPF from laboratory studies, field observations under various environments, and NPF parameterizations in atmospheric models are discussed in sections 3–5, respectively. Section 6 presents an overview on the outstanding questions, challenges, and directions for atmospheric NPF research. Appendix A contains a list of acronyms.

There are several reviews on the topic of NPF from different aspects. The Kulmala, Vehkamäki, et al. (2004) review is an earliest overview of NPF observations made at various locations. R. Zhang et al. (2012) provide a comprehensive review of theories and observations of aerosol nucleation and growth. Hirsikko et al. (2011) review air ion measurements. Theoretical aspects of homogeneous nucleation are discussed in several overview papers (Curtius, 2006; Enghoff & Svensmark, 2008; Hegg & Baker, 2009; Kazil et al., 2008; Vehkamäki et al., 2012). Kulmala, Laakso, et al. (2014) discuss the instrument development related to NPF. More recently, Yu, Ren, and Kanawade (2017), Z. Wang et al. (2017), and Chu et al. (2019) discuss NPF

occurring under extremely polluted conditions. Kerminen et al. (2018) review recent findings of NPF from field observations in different atmospheric regions. Semeniuk and Dastoor (2018) discuss NPF parameterizations used in regional air quality models. The present review focuses on the latest progress in development of new instruments, laboratory observations of multicomponent nucleation, and field observations of NPF at under various ambient conditions and discusses the critical needs for the development of physically based NPF parameterizations in atmospheric models.

2. Development of New Instruments

Measuring aerosol sizes, especially those including newly formed clusters, is necessary for NPF studies but very challenging. Great progress has been made recently in this direction. Chemical analysis of nucleation precursors is extremely important, as nucleation is a nonlinear process. In this section, we discuss analytical methods used to detect major nucleation precursors, sulfuric acid, ammonia and amines, and newly nucleated gas phase clusters, both charged and neutral.

2.1. Sizing of Nanoparticles

Understanding NPF requires reliable instrumentation capable of the detection of forming clusters as well as their precursors. Measurement of particle concentrations and size distributions down to a few nanometers, preferably down to ~1.5-nm mobility diameter (Kulmala et al., 2012), is especially important, because this information is needed to identify NPF events and retrieve parameters describing particle formation processes, for example, formation and growth rates (*GR*).

The most common particle size spectrometer is Differential or Scanning Mobility Particle Sizer (D/SMPS), which consists of a neutralizer, a Differential Mobility Analyzer (DMA), and a particle counter. D/SMPS have been widely used in atmospheric measurements, but large deviations still exist in individual instrument performances especially in the sub-10-nm size range (Wiedensohler et al., 2012). Much progress has been thus made in recent years to develop instruments optimized for the measurement of smallest size particles (e.g., down to ~1 nm).

High-Resolution DMA (HR-DMA) was developed to accurately size particles and clusters down to the molecular sizes. Several studies characterized the performance of HR-DMAs down to ~1 nm (Brunelli et al., 2009; J. K. Jiang et al., 2011; Kangasluoma et al., 2016; Steiner et al., 2010; M. R. Stolzenburg et al., 2018). For atmospheric measurements, sufficiently high transmission of small particles through the DMA is required, since concentrations of these small particles are generally low.

An important factor limiting the size range in a particle spectrometer is lower activation efficiencies of Condensation Particle Counters (CPCs) at smaller sizes. The minimum size of water- or butanol-based CPCs is typically ~2–3 nm before they are homogenously nucleated by the working fluid. Instruments using diethylene glycol (DEG) as the working fluid can reach even smaller sizes without homogenous nucleation (Iida et al., 2009). Several particle counters using DEG have been developed (Kuang et al., 2012; Vanhanen et al., 2011), but all of them require a second stage growth chamber, where small particles grow to the optical sizes to be detected by light scattering.

More recent particle size spectrometers that combine HR-DMAs and low cutoff particle counters can detect particle sizes smaller than 2 nm. These instruments include D/SMPS systems with DEG-based CPC (J. Jiang et al., 2011; Kangasluoma et al., 2018), as well as the DMA-train (Stolzenburg et al., 2017) that consists of several DMAs in parallel set to different cutoff sizes. The main disadvantage of these new instruments is low counting efficiencies of particles at small sizes, due to low charging probabilities, low transmission efficiencies through the DMA, and diffusion losses throughout the system.

Other types of size spectrometers include Air Ion Spectrometer (AIS) that detects naturally charged ions using electrometer (Mirme & Mirme, 2013). Neutral cluster and Air Ion Spectrometer (NAIS) can measure particle size distributions of total aerosol particles (including charged and neutral) above ~2 nm, using corona discharge (Mirme & Mirme, 2013). Ion spectrometers often use large flow rates to reduce particle losses, but the associated electric noise can be an issue at low particle concentrations.

NPF has also been studied using CPC-based methods to retrieve the particle size information. Although less accurate in sizing than DMA-based methods, these instruments have an advantage in generating more

signals at low concentrations of particles. The scanning Particle Size Magnifier (PSM, Vanhanen et al., 2011) has been used to inversely derive particle size information down to ~1 nm, by continuously changing the supersaturation ratio of the DEG working fluid to activate a different size fraction of sub-3-nm particles at a time (Kangasluoma et al., 2015; Lehtipalo et al., 2014). Another approach is to use pulse-height analysis to resolve the original particle sizes based on measured droplet size distributions (Kuang, 2018; Sipila et al., 2009). The main limitation of both aforementioned methods is that activation of particles inside the CPC is sensitive to chemical composition of particles (Kangasluoma et al., 2014; Wimmer et al., 2013), so for atmospheric aerosols with unknown chemical compositions, this issue can make analysis more challenging.

2.2. Chemical Analysis of Nucleation Precursors

Atmospheric observations and laboratory studies of NPF have shown that Chemical Ionization Mass Spectrometer (CIMS) is a powerful tool for sulfuric acid measurements. The CIMS is based on the ion-molecular reaction using nitrate ions (NO_3) as reagent (Eisele & Tanner, 1993) at atmospheric pressure. In general, the detection limit of CIMS is at $10^5 \, cm^{-3}$ or even lower, with an integration time of 1 min, with the overall uncertainty of ~60% (Benson et al., 2008; Benson et al., 2009; Eisele & Tanner, 1993; Erupe et al., 2010; Kürten et al., 2012; Petäjä et al., 2009).

Nitrate ions (and their higher order clusters) are produced in a flow annular to the sample flow by exposing a parts per million by volume mixture ppmv of nitric acid in air to a commercial X-ray source (<10 KeV) or a ^{210}Po radiation source. During this process, hydroxyl (OH) radicals can also form from water molecules by the ionization source, and if they react with SO_2, they can produce artifact sulfuric acid molecules. To reduce such artifacts, an electric field is applied in the ion-molecule reaction region so that only charged nitrate ions, as opposed to OH radicals, flow in to the reaction region to react with sulfuric acid molecules (Eisele & Tanner, 1993). Calibration of sulfuric acid is made based on the known concentrations of OH, produced from ultraviolet photolysis of water molecules, by converting the OH radicals to sulfuric acid in the presence of excessive SO_2 concentrations (Kürten et al., 2012; Sjostedt et al., 2007; Tanner & Eisele, 1995; Young et al., 2008).

It is possible that strong base compounds, by clustering with sulfuric acid, can affect the detection of sulfuric acid in CIMS (Kurten et al., 2011). However, Rondo et al. (2016) recently investigated the effects of amine-sulfuric acid clustering on the detection of sulfuric acid with CIMS and concluded that the nitrate-CIMS can measure sulfuric acid monomers efficiently even in the presence of amines.

An atmospheric pressure ionization-ion drift-CIMS was developed to measure ambient sulfuric acid and its clusters (Zheng et al., 2010; Zheng et al., 2011). The application of the drift tube facilitates controlled ion-molecule reactions, with the advantages to detect and quantify gases without the necessity of calibrations using authentic standards and to effectively prevent unwanted cluster formation (Fortner et al., 2004; Ji et al., 2017; Zhao et al., 2004; Zhao et al., 2005; Zhao & Zhang, 2004). The atmospheric pressure ionization-ion drift-CIMS exhibits the advantages over the low pressure ion drift-CIMS and atmospheric pressure ionization-CIMS, by improving the detection sensitivity by 3 orders of magnitude and a factor of 3, respectively (Zheng et al., 2010).

Amines have been measured with offline analytical methods based on gas chromatography, high-performance liquid chromatography, and ion chromatography, coupled with flame ionization detectors, mass spectrometry, ultraviolet detectors, fluorescence detector, or electrochemical detectors (Ge et al., 2010). The advantage of these offline techniques is that the detection limit can be improved by an enhanced analyte preconcentration (e.g., absorption in water or adsorption on solid sorbents), but the preconcentration with the prolonged sampling time also raises several technical issues. High-performance liquid chromatography and ion chromatography methods often involve amine absorption in water; and water contains amines as impurities (Erupe et al., 2011; Yu, McGraw, & Lee, 2012). Nevertheless, DMA has been measured by the ion chromatography method in CLOUD chamber experiments with an extremely low detection limit at the sub-parts per trillion by volume (pptv) level (Praplan et al., 2012).

CIMS have been used for fast-time resolution detection of various atmospheric amines and ammonia (Eisele, 1988; Hanson et al., 2011; Jen, Bachman, et al., 2016; Kürten, Bergen, et al., 2016; Nowak et al., 2010; Sellegri et al., 2005; Simon et al., 2016; You et al., 2014). Hanson et al. (2011) and Zheng et al. (2015) have used

protonated water ions as reagent to measure various amines and ammonia. H. Yu and Lee (2012) applied protonated ethanol and acetone ions as reagent to detect amines and ammonia together, and this ion-molecule reaction scheme later was applied for amine measurements also by other groups (Yao et al., 2016; Yao et al., 2018). Either quadrupole mass spectrometry or high-resolution time of flight mass spectrometer (HR-TOF-MS) has been used as detector. These CIMS can provide detection limits of pptv or sub-pptv for amines and several tens pptv of ammonia, with the time resolution of 1 min (You et al., 2014).

The main challenge of measuring amines and ammonia is to make reliable background measurements to ensure the measured signals truly represent the concentrations of amines and ammonia. Background measurements should "shield out" the effects of artifact signals rising from the desorption of amines and ammonia from the sampling inlets and the effects of ambient conditions such as relative humidity (RH) and temperatures and other atmospheric species on ammonia and amine signals (Hanson et al., 2011; You et al., 2014; Yu & Lee, 2012). Thus, the zero air used in background measurements should be different from the zero air typically generated from a commercial "zero air generator," because the latter can show significantly lower background signals, and thus, measurements can show *higher* concentrations of amines than the actual values. Also, because sensitivities are dependent on the background signal levels, frequent in situ calibrations are needed (You et al., 2014).

Reliable measurements of gas phase precursors such as sulfuric acid, organics, ammonia, and amines with sufficiently low detection limits and high time response are needed for future studies. Measurements should provide the robust calibration and stability during the long-term operation and be reproducible under different atmospheric conditions with varying temperatures, RH, and air pressures.

2.3. Chemical Analysis of Charged and Neutral Clusters

Prenucleated clusters containing sulfuric acid have been measured by CIMS (Eisele et al., 2006; Froyd & Lovejoy, 2003; Froyd & Lovejoy, 2004; Hanson & Eisele, 2000; Zhao et al., 2010; Zhao et al., 2011). Recently, chemical composition of atmospheric clusters (e.g., beyond tetramers) has been measured with Atmospheric Pressure ionization-Time of Flight mass spectrometer (APi-TOF; Junninen et al., 2010) and Chemical Ionization-Atmospheric Pressure Ionization-Time of Flight mass spectrometer (CI-APi-TOF; Jokinen et al., 2012), both based on the HR-TOF-MS technique. The APi-TOF is in fact an HR-TOF-MS which is attached to an Atmospheric Pressure interface (APi). In this case, no chemical ionization is applied to the sample flow, and it detects ions that naturally form in the atmosphere. The APi-TOF can be configured to measure either positive or negative ions.

The use of the HR-TOF-MS improves ion transmission by up to 2 orders of magnitude over a typical quadrupole mass spectrometry operated in m/z scanning mode. Complete mass spectra can be acquired at rates greater than 100 Hz with mass resolving power between 3,000 and 7,000 m/z and mass accuracy better than 10 ppm. The nitrate CI-APi-TOF detects sulfuric acid, aminium, ammonium, iodic acid, and HOMs (Jokinen et al., 2012; Kürten, Bergen, et al., 2016; Sipilä et al., 2016). The CI source for the nitrate CI-APi-TOF is based on the Eisele and Tanner (1993) described in section 2.2. The quantification of HOM concentrations has been achieved using the same calibration factor for the sulfuric acid detection, under the assumption that HOMs are detected near the collision limit and have the same transmission efficiencies as sulfuric acid. The lack of direct calibration and peak identifications of HOM species is the major technical issue for CI-APi-TOF. Figure 3 shows mass spectra taken by CI-APi-TOF during α-pinene ozonolysis reactions in the Jülich Plant Atmosphere chamber, indicating a series of monomers and dimers of HOMs formed from biogenic VOC (BVOC) ozonolysis reactions (Bertram et al., 2011; Brophy & Farmer, 2015; Ehn et al., 2014; Veres et al., 2008).

Other ion chemistries are also applied to CI-APi-TOF to measure specific classes of organic compounds, using a low-pressure ion molecule reactor. Each chemical ionization method has its own advantages and disadvantages. The advantage of nitrate anion is that it is selective and relatively easy to interpret the complex spectra. The acetate anion can detect carboxylic acids (Brophy & Farmer, 2015; Veres et al., 2008). However, there is acetate anion clustering and significant fragmentation including dehydration. Iodide anion clustering is similar to nitrate except that it can see less oxidized species (Bertram et al., 2011). It has the same advantage as the nitrate chemical ionization when looking only at iodide-containing clusters that are easily resolved in the spectra.
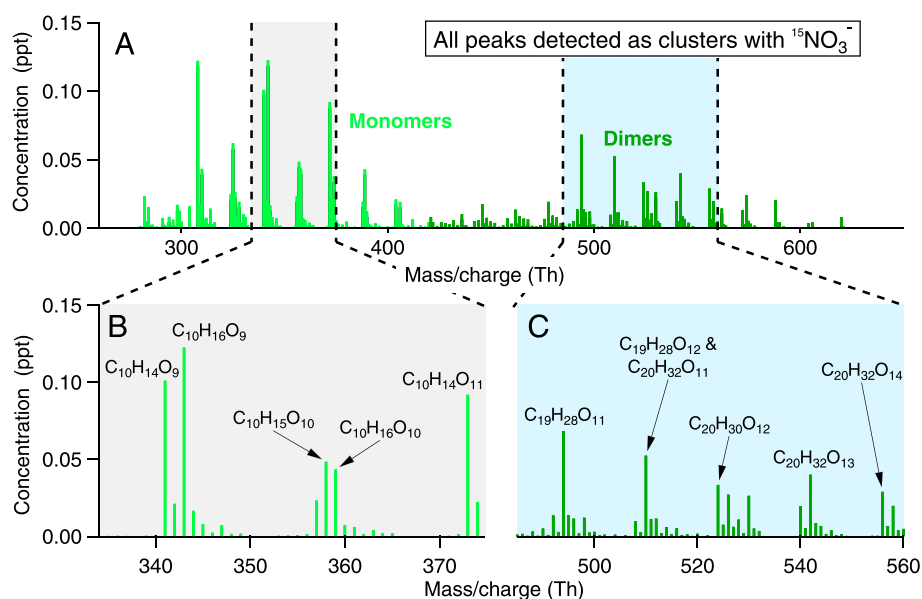
Figure 3. Mass spectra taken by APi-TOF during α-pinene ozonolysis reactions in the Jülich Plant Atmosphere chamber. Adapted from Ehn et al. (2014).

The large variety of oxidized organic compounds poses specific challenges for instrumentation, as different ion chemistries used in the CIMS instruments measure different subgroups of organic compounds. For completeness, different techniques and ion chemistries should be used in parallel, ideally, but it is not straightforward to integrate or intercompare the mass spectrometer data obtained from different instruments. Also, the current mass spectrometers still suffer from fragmentation and thermal decomposition of clusters and particles during the sampling and measurement, which hinders the accurate quantification of organic species.

With the recent instrumentation development, we are now able to measure the chemical composition (e.g., molecular mass, although without chemical identities) of newly nucleated clusters (Jokinen et al., 2012; Junninen et al., 2010). There are several instruments that were designed to measure chemical composition of sub-20-nm particles (Gomez-Hernandez, 2015; Horan et al., 2017; Smith et al., 2004), but these instruments have been primarily used for particles larger than ~20 nm. Thus far, chemical analytical techniques that cover the particle size range from ~3 to 20 nm are mostly missing, because these particles are difficult to ionize, have extremely low mass concentrations, and are very diffusive. There have been some indirect methods used for chemical analysis of this size range particles, for example, Tandem DMA (Laaksonen et al., 2008; Petäjä et al., 2005). Understanding the chemical composition of particles from 3 to 20 nm particles is essential for understanding the growth mechanisms of new particles and their effects on CCN formation.

Atmospheric sensors currently used for chemical analysis and size distribution measurements are very costly and labor consuming to operate and troubleshoot. Due to their high costs, deployment of these instruments at a wide range of atmospheric locations, especially in developing countries and remote locations, is not feasible. Due to various technical issues in their operation, they are usually not suitable for long-term, online measurements, especially at mountain-top sites and onboard the aircraft. Development of low-cost, portable, and reliable instruments that can provide detailed size information of clusters and nanoparticles is needed.

3. Nucleation and Growth Mechanisms: New Results From Laboratory Studies

Laboratory nucleation observations conducted under well-controlled experimental conditions are needed to test and constrain nucleation theories, but nucleation experiments are extremely difficult to perform. The major technical challenges include difficulties in measuring very low concentrations of nucleation precursors and the chemical composition of newly formed particles in the size range from the critical clusters

(~1.5 nm) up to ~10-nm particles. Experiments often suffer various contaminations of nucleation precursors, especially relatively high concentrations of amines and ammonia that originate from deionized water. For example, flow tubes (e.g., made by Pyrex) typically contain the pptv and tens pptv level of base compounds depending on RH, even after very deliberate cleaning processes (Erupe et al., 2011; Yu, Dai, et al., 2017; Yu, McGraw, & Lee, 2012), with some exceptions (Glasoe et al., 2015; Hanson et al., 2017). As nucleation is a nonlinear process and the J are extremely sensitive to the temperature, RH, and concentrations of nucleation precursors, it is difficult to obtain consistent data from different experimental setups. The chamber experiment results are also difficult to interpret, because wall loss of nucleation vapors (Pierce et al., 2008; Zhang et al., 2014), clusters (Ehrhart & Curtius, 2013; Kürten et al., 2015; McMurry & Li, 2017), and particles (Pierce et al., 2008) must be accounted for the accurate calculation of J and GR. Nucleation studies have been made with fast flow reactors (Ball et al., 1999; Benson et al., 2011; Berndt et al., 2010; Brus et al., 2010; Hanson & Lovejoy, 2006; Lovejoy et al., 2004; Young et al., 2008; Zhang et al., 2004), expansion cloud chambers (Katz, 1970), environmental smog chambers (Metzger et al., 2010; Riccobono et al., 2012), real plant chambers (Hao et al., 2009; Joutsensaari et al., 2005; Mentel et al., 2009), and recently at CERN in the large environmental chamber CLOUD (Almeida et al., 2013; Kirkby et al., 2016; Kürten et al., 2014; Lehtipalo et al., 2016; Tröstl et al., 2016). The advantage of CLOUD is that it has extremely low concentrations of impurities and is equipped with a large number of state-of-the-art aerosol and chemical instruments through collaborations. Even the CLOUD chamber still suffers from the contamination of base compounds at high RH and high temperature conditions due to desorption of base compounds from the stainless steel walls (Kürten, Bianchi, et al., 2016). In this section, we highlight key scientific findings from the recent laboratory experiments of nucleation and early growth.

3.1. Base-Stabilization Nucleation

Sulfuric acid is the most important atmospheric nucleation precursor, and base compounds such as ammonia and amines also enhance nucleation of sulfuric acid. This is because base compounds can reduce the vapor pressure of sulfuric acid and thus stabilize critical clusters. Low molecular mass alkyl amines were shown to be stronger stabilizing agents for sulfuric acid clusters than ammonia by laboratory experiments and quantum chemistry (Jen et al., 2014; Kurtén et al., 2008; Yu, McGraw, & Lee, 2012).

Ammonia and amines have been observed in small sulfuric acid clusters in both the laboratory and the atmosphere (Almeida et al., 2013; Bianchi et al., 2014; Hanson & Eisele, 2002; Kirkby et al., 2011; Kürten et al., 2014; Schobesberger et al., 2015; Yao et al., 2018; Yu, McGraw, & Lee, 2012). Figure 4 shows an approximate 1:1 molar ratio of sulfuric acid and ammonia (or DMA) in newly formed clusters at the atmospherically relevant gaseous [NH$_3$]/[H$_2$SO$_4$] concentration ratios of 10–500 (Bianchi et al., 2014; Schobesberger et al., 2015). While amines can strongly enhance the J (Almeida et al., 2013; Glasoe et al., 2015), studies show there is a saturation effect of J or dimer concentrations with a further increase in DMA concentrations, for example, when exceeding 5 pptv DMA at temperatures between 278 and 300 K (Almeida et al., 2013; Chen et al., 2012; Jen et al., 2014; Kürten et al., 2014). With 5–32 pptv or higher concentrations of DMA, formation of neutral sulfuric acid dimers reaches the kinetic limit with a power dependence of J on [H$_2$SO$_4$] of 2, indicating highly stable clusters with negligible evaporation (Almeida et al., 2013; Kürten et al., 2014). This is equivalent to the notion that the critical cluster size is smaller than the dimer.

It should be noted, however, that these laboratory observations do not necessarily mean that nucleation always proceeds at the kinetic limit in the real atmosphere, because nucleation pathways depend on the concentration as well as the stabilizing ability of base compounds. For example, Jen et al. (2014) found that the saturation concentration with respect to sulfuric acid dimer concentration was 1,800–2,300 pptv for ammonia, in comparison to ~20 pptv in the case of DMA. M. Chen et al. (2012) observed that the measured sulfuric acid tetramer concentrations in Atlanta and Mexico City were approximately equal to the collision-limited values, but the measured trimer and dimer concentrations were a factor of 8 and 16 below the collision-limited values, respectively. This implies that monomer evaporation from the dimer and trimer is the bottleneck for nucleation under these specific conditions. In Chinese megacities, average [NH$_3$] of 1.7 ppbv were reported in Nanjing, and total C1–C6 amines of ~79 pptv were reported in Shanghai (Yao et al., 2016; Zheng et al., 2015). It is thus very likely that nucleation would proceed at the collision-limit of sulfuric acid in these highly polluted environments. Figure 2 shows the J and sulfuric acid concentrations measured in urban Shanghai, China (Yao et al., 2018) and compares them with the CLOUD DMA-ternary nucleation results
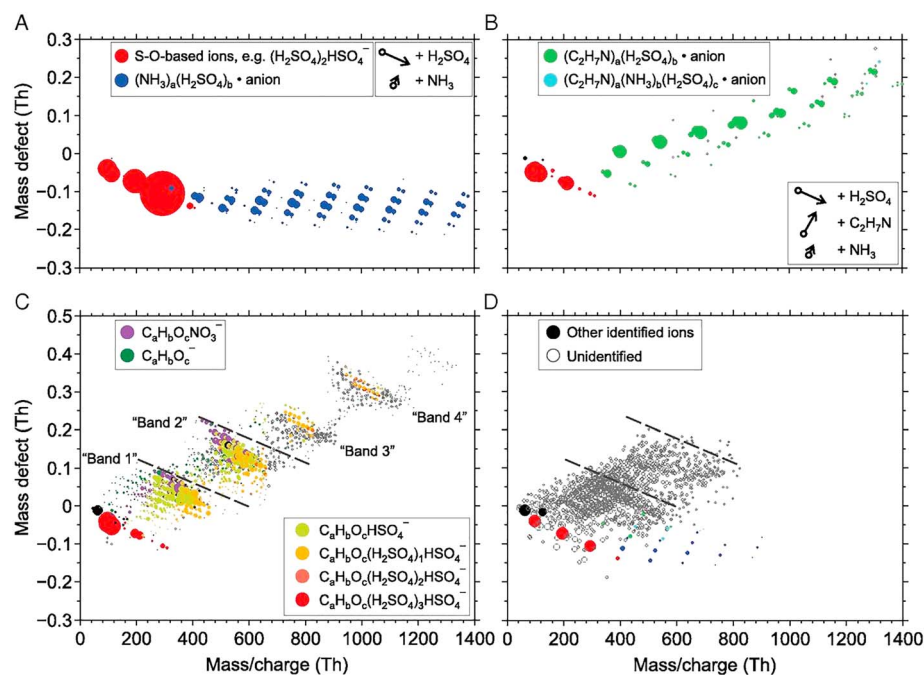
Figure 4. Api-TOF mass defect diagram for negatively charged clusters during new particle formation experiments at CLOUD chamber (a–c) and in the boreal forest (d). The results from experiments of ternary nucleation with sulfuric acid and ammonia (a), ternary nucleation with DMA (b), and multicomponent nucleation with sulfuric acid and oxidation products of pinanediol (c) are shown. Colors indicate different ions, and circle diameters are proportional to ion counts. Adapted from Schobesberger et al. (2013).

(Almeida et al., 2013). These results imply that base compounds in these very polluted cities can be important for aerosol nucleation, but the effects of other chemical species such as biogenic and anthropogenic organics also cannot be excluded.

Amines also have an important effect on *GR* of new particles. Recently, Lehtipalo et al. (2016) observed that when 5 to 70-pptv DMA was present in the CLOUD chamber, the *GR* of 2 nm particles was a factor of ~10 higher than that could be expected based on the mass flux calculated from the apparent sulfuric acid concentrations and cocondensation of DMA. It was thus concluded that the dominant growth mechanism is a *cluster-cluster* collision process, instead of sulfuric acid *monomer-cluster* collision. This finding has an important atmospheric implications especially in a highly polluted atmosphere. In the past, the higher *GR* could not be explained by sulfuric acid condensation and were often attributed to additional contribution from some unidentified organics (Yu, Ren, & Kanawade, 2017).

Laboratory observations show that the enhancement of nucleation is dependent on the proton affinity of base compounds (Berndt et al., 2014; Glasoe et al., 2015; Yu, McGraw, & Lee, 2012). The enhancement of nucleation is also stronger by ammonia and an amine together, compared to ammonia or an amine compound alone (Berndt et al., 2014; Glasoe et al., 2015; Yu, McGraw, & Lee, 2012). Considering many common emission sources (Ge et al., 2010) and the copresence of amines and ammonia in the atmosphere (You et al., 2014; Yu & Lee, 2012), such synergetic effects have important atmospheric implications. Systematic laboratory studies are needed to understand the role of different amines and the synergetic effects on sulfuric acid nucleation. Amines undergo rapid oxidation reactions in the atmosphere (Finlayson-Pitts & Pitts, 2000). Studies have shown that some oxidation products such as imines are very efficient in aerosol nucleation and secondary organic aerosol (SOA) formation (Nielsen et al., 2011). Future studies are required to understand how oxidation products of amines affect aerosol nucleation and growth.

3.2. HOMs Formed From VOCs Oxidation Reactions

Although sulfuric acid and bases are known to be the key precursors for NPF, sulfuric acid alone, or even together with base compounds, cannot explain atmospheric observations in most part of the continental

boundary layer. The importance of organic vapors in particle formation, and most certainly in particle growth, has long been discussed (e.g., Riipinen et al., 2012, and references therein). R. Zhang et al. (2004) for the first time showed that organic compounds such as benzoic acid can enhance sulfuric acid nucleation, due to hydrogen bonding between sulfuric acid and benzoic acid molecules. Metzger et al. (2010) showed trimethylbenzene (in the absence of ozone or OH) enhances sulfuric acid nucleation.

For organic compounds to participate in particle formation and in the early stage of growth, they need to have extremely low saturation vapor pressures as well as high enough concentrations to overcome the Kelvin barrier and condense on nanometer-sized aerosol particles or form clusters by themselves. Saturation vapor pressures of oxidation products of VOCs span over many orders of magnitude and can be classified into the VOC, intermediate VOC, semi-VOC (SVOC), low volatility organic compound (LVOC), and extremely low VOC (ELVOC) (Donahue et al., 2011). For example, log saturation vapor concentrations (C^* in $\mu g/m^{-3}$) of ELVOC, LVOC, and SVOC may range approximately from -10 to 3 (Schobesberger et al., 2013). ELVOC and LVOC together are usually denoted broadly as HOMs, although there are some cases where (E)LVOC and HOMs are not necessarily the same (Bianchi et al., 2019).

HOMs are produced from organic peroxides formed from oxidation reactions of BVOCs and anthropogenic VOCs (AVOCs), via autoxidation reactions involving successive intramolecular H-shift followed by O_2 addition (Berndt et al., 2016; Crounse et al., 2011; Crounse et al., 2013; Ehn et al., 2014; Ehn et al., 2017). HOMs contain a wide range of chemical functional groups such as peroxides, hydroperoxides, carbonyls, and percarboxylic acids. Due to their high O/C ratios (close to unity), they have sufficiently low volatilities to contribute to aerosol nucleation and growth. Several studies have quantified HOM yields from monoterpenes (Ehn et al., 2014; Ehn et al., 2017; Jokinen et al., 2015), isoprene (Jokinen et al., 2015), and AVOCs (e.g., alkyl benzenes; Molteni et al., 2018; Praske et al., 2018; S. Wang et al., 2017). The exact formation pathways and properties of HOMs are still under investigation (Berndt et al., 2016; Bianchi et al., 2019; Ehn et al., 2017).

Schobesberger et al. (2013) have shown that HOMs (e.g., $C_{10}H_{12-16}O_{2-10}$) formed from OH oxidation reactions of pinanediol ($C_{10}H_{18}O_2$, a first generation oxidation product of monoterpenes) interact with sulfuric acid to form monomers to tetramers (Figure 4), and these HOMs can enhance J for sulfuric acid. Subsequently, Riccobono et al. (2014) derived that J in the same nucleation system are dependent on sulfuric acid with the second power and on HOMs linearly. Ehn et al. (2014) have demonstrated the formation of HOMs from ozonolysis of α-pinene and derived that the J are dependent on sulfuric acid linearly and on HOMs with the second power. They also showed that growth of particles in the size range between 5 and 50 nm can be explained exclusively with ELVOC (without LVOCs and SVOCs). On the other hand, the flow tube experiments by Berndt et al. (2014) showed that HOMs produced from ozonolysis of monoterpenes do not enhance sulfuric acid nucleation. Kirkby et al. (2016) claimed that nucleation can proceed from HOMs (from α-pinene ozonolysis) alone, assisted by ions. Tröstl et al. (2016) showed that *GR* of 5 to 50 nm particles cannot be explained by HOMs alone, suggesting that LVOCs that are not measured by the nitrate-CIMS and some additional SVOCs may be also involved in the growth. Hao et al. (2009) have shown that oxidation of real plant-derived VOCs by OH can produce very different nucleation and growth rates than oxidation by O_3. These discrepancies imply that HOMs formed from different precursors and different pathways under different conditions (e.g., different VOCs and different oxidants such as O_3 vs. OH vs. NO_3) can contribute to aerosol nucleation and growth in different ways.

Recently, Lehtipalo et al. (2018) and McFiggans et al. (2019) showed that the exact mixture of precursors is crucial in determining particle formation rates in the systems involving HOMs and other compounds. But how AVOCs and BVOCs interact together to affect aerosol nucleation and growth has not been investigated. This is especially relevant in extremely polluted regions in the tropics and subtropics where there are also strong biogenic emissions of BVOCs. Understanding these multicomponent nucleation systems of mixed chemical species requires comprehensive instrumentation and coordinated laboratory and filed studies.

Under high NO_x conditions, there is a competition between autoxidation and RO_2-NO_x reactions (which leads to formation of organonitrate HOMs, HOM-ONs). Since NO_x concentrations are abundant in large parts of the daytime boundary layer, especially in urban environments, it is necessary to understand how NO_x affect nucleation and growth from HOMs. Chamber studies showed that NO_x can suppress biogenic NPF from terpenes (Lehtipalo et al., 2018; Wildt et al., 2014). Wildt et al. (2014) studied the impact of NO_x

on NPF and on photochemical ozone production from the real plant BVOC emissions. At high NO_x conditions (e.g., [BVOC]/[NO_x] < 7 and [NO_x] > 23 ppb), NPF was suppressed. Instead, photochemical ozone formation was observed at higher OH and lower NO_x concentrations. At the same time, NO_x had little effect on the mass concentration of SOA. Other studies, on the other hand, indicate a strong increase in biogenic SOA due to NO_x (e.g., Shrivastava et al., 2019). It is possible that organonitrate HOMs (HOM-ONs), which are formed from biogenic vapors in the presence of NO_x, are much less efficient in forming particles than other HOMs (Lehtipalo et al., 2018), probably due to their higher volatilities, but they might still be efficient in the aerosol growth. Future studies are required to understand the relative roles of HOMs versus HOM-ONs on aerosol nucleation and growth.

3.3. Stabilizing Ions

Ions can enhance nucleation, especially when neutral nucleation is weak. This is because charged clusters coagulate faster than neutral clusters and in turn stabilize the critical clusters and reduce the Gibbs free-energy barrier for cluster formation (Seinfeld & Pandis, 2016). Researchers have used different terms for ion nucleation, such as ion-induced nucleation (IIN; Laakso et al., 2004; Lovejoy et al., 2004; Modgil et al., 2005) and ion-mediated nucleation (IMN; Yu, 2010; Yu & Luo, 2014b; Yu & Turco, 2000; Yu & Turco, 2001; Yu et al., 2008), but in this review we use a more general term "ion nucleation" to include both IIN and IMN.

Earlier ion nucleation studies were performed in the laboratory and showed in general that J are enhanced by charges (Kim et al., 1997; Wilhelm et al., 2004). Lovejoy and colleagues performed flow tube experiments of ion nucleation for the binary homogeneous nucleation of sulfuric acid and water system (Froyd & Lovejoy, 2003; Froyd & Lovejoy, 2004; Lovejoy et al., 2004). They measured the thermodynamics for growth and evaporation of prenucleated sulfuric acid cluster ions and incorporated these data into a kinetic aerosol model to yield quantitative predictions of the rate of ion nucleation. This IIN parameterization was tested and constrained by the temperature, RH, SO_2, OH and H_2SO_4 concentrations, and aerosol size distributions measured in a wide range of conditions from the ground level to the upper troposphere and lower stratosphere (Lee et al., 2003), which enabled the model to parameterize the atmospheric conditions using more realistic parameters for atmospheric nucleation. The model predicts that the negative ion nucleation is more important than positive ion nucleation and that ion nucleation of sulfuric acid and water mechanism is an efficient source of new particles in the middle and upper troposphere. However, the model does not generally predict the nucleation events observed in the boundary layer.

The recent CLOUD measurements quantified the enhancement of ion nucleation compared with neutral nucleation for several chemical systems (Kirkby et al., 2011). For the sulfuric acid and water binary nucleation, ions are more important at higher atmospheric temperatures than lower temperatures (Duplissy et al., 2016). For the ammonia-ternary nucleation, the contribution of ion nucleation is small at temperatures between 208 and 248 K when ammonia is present at several pptv or higher, but the presence of ions significantly enhances J for higher temperatures independent of ammonia levels (Kürten et al., 2016). Ion nucleation of sulfuric acid and ammonia was recently suggested to drive NPF in the coastal Antarctica (Jokinen et al., 2018).

For the case of ternary sulfuric acid and DMA, the contribution of ion nucleation is generally small, reflecting the high stability of sulfuric acid-DMA clusters and indicating that galactic cosmic rays exert only a small influence on their formation (Almeida et al., 2013). Ion nucleation is also less important when HOMs are mixed with sulfuric acid and ammonia (Lehtipalo et al., 2018; Riccobono et al., 2014). The current conclusion from the CLOUD experiments is that although ions can be involved in nucleation in large parts of the atmosphere, the dependence on ion concentrations and thus variation in ionization rates are weak (Dunne et al., 2016).

Kirkby et al. (2016) showed that ions formed from galactic cosmic rays at the ground level ionization rates can increase J by 1 to 2 orders of magnitude compared with neutral nucleation for the HOMs produced from monoterpene ozonolysis reactions. Based on this study, modeling studies have suggested that pure biogenic ion nucleation process can be an important contributor to the CCN production in preindustrial terrestrial environments (Gordon et al., 2016). However, the role of pure biogenic ion nucleation in present day pristine environments, such as in Amazon rainforests, is still high uncertain, because atmospheric observations

showed that NPF is entirely absent in these pristine forests even with strong emissions of BVOCs (Rizzo et al., 2018).

3.4. Temperature and RH Dependencies

Temperature and RH are the key thermodynamics parameters of aerosol nucleation and growth (Seinfeld & Pandis, 2016). J is a function of Gibbs free-energy barrier and temperature. At lower temperatures, the Gibbs free-energy barrier is lowered, and the critical cluster becomes smaller. RH is also the saturation ratio and the chemical activity of water. In the atmosphere, temperature and RH vary greatly depending on altitude, latitude, season, and the time of the day. Therefore, to simulate NPF correctly in the atmosphere, both J and GR as a function of temperature and RH should be understood.

Duplissy et al. (2016) conducted binary homogeneous nucleation of sulfuric acid and water, with and without ions in the CLOUD chamber, at temperatures from 207 to 299 K, and RH between 11% and 58%. At lower temperatures, both ion nucleation and neutral binary nucleation are at the kinetic regime, while at higher temperatures, J is strongly dependent on [H_2SO_4], indicating there are high Gibbs free-energy barriers. Kürten, Bianchi et al. (2016) showed the temperature dependence of ternary nucleation in the CLOUD chamber, at conditions with temperatures from 208 to 298 K, [H_2SO_4] between 10^5 and 10^9 cm^{-3}, and ammonia mixing ratios up to ~1,400 pptv. At 208 K, J exceeded the threshold of 1 cm^{-3} s^{-1} at a [H_2SO_4] of ~3×10^6 cm^{-3} for the binary case and at [H_2SO_4] of ~5×10^5 cm^{-3} for the ternary case with 5 pptv of ammonia. Kürten, Bianchi et al. (2016) showed that ternary nucleation involving ammonia could be an important mechanism for nucleation in the polluted boundary layer when temperatures are not too high (e.g., 278 K).

Yu, Dai, et al. (2017) performed flow tube experiments of sulfuric acid aerosol nucleation, at temperatures ranging from 248 to 313 K, RH from 1% to 79%, and under minimal base concentrations (ammonia < 23 pptv, methylamine < 1.5 pptv, and DMA < 0.52 pptv). Yu, Dai, et al. (2017) provided for the first time the temperature and RH dependence of both J and GR. J shows the following dependence within the experimental conditions:

$$J = 10^{41.8}[RA]^3[RH]e^{\frac{-2.4 \times 10^4}{T}} \tag{7}$$

where RA is the saturation ratio (or relatively acidity) of sulfuric acid. GR is independent of temperature below 290 K but significantly decreases at temperature above 290 K, while RH has a moderate effect on GR.

Recent studies have shown that oxygenated organics formed from BVOCs can grow newly nucleated particles in a wide range of troposphere temperatures (D. Stolzenburg et al., 2018). This is because gas phase autoxidation reactions involved in the formation of HOMs usually have strong temperature dependencies, with higher reaction rates at higher temperatures (Frege et al., 2018), whereas nucleation is favored at lower temperatures. Despite the low reaction rates, Bianchi et al. (2016) observed high concentrations of HOMs during NPF at a high-elevation site with low temperatures, with an average temperature of −5 °C at noontime. At present, however, there is still no clear evidence from atmospheric observations which demonstrates "causal effects" of temperature and RH on NPF.

3.5. Thermodynamics Measurements

Perhaps the most significant way to improve CNT theories is to directly measure Gibbs free-energy changes in cluster formation and surface tension and density of newly formed particles. Calculations have shown that even small changes in thermodynamics can result in significant changes in cluster distributions (Hanson et al., 2017). In this context, the most important future work will be to directly measure Gibbs free-energy changes of aerosol nucleation. Hanson and Lovejoy (2006) and Froyd and Lovejoy (2003, 2004) have conducted thermodynamics measurements of prenucleated sulfuric acid neutral and charged clusters. Since the concentrations of prenucleated and nucleated clusters are extremely low and their lifetimes are very short, the measurement of Gibbs free-energy changes from monomer to critical clusters is extremely challenging. Also measurements are needed to detect surface tension and densities of nucleated clusters, as well as the effects of organics on the surface tension of sulfuric acid clusters, is another challenging area for future work.

4. NPF Observations in Various Environments

Hundreds of measurements have been made over the world during the past decades on various platforms, such as at ground level, on aircraft, and at high mountain sites (Chu et al., 2019; Kerminen et al., 2018; Kulmala, Vehkamäki, et al., 2004; Z. Wang et al., 2017; Yu, Ren, & Kanawade, 2017; Zhang et al., 2012). Measurement locations varied from clean rural sites to polluted urban sites to extremely polluted megacities, from tropics to polar regions, and from coastal sites to forested regions. These measurements in general show a distinctive seasonality with higher NPF frequency in spring and higher GR during the summer, with some exceptions (Nieminen et al., 2018). These observations show quite conclusively that sulfuric acid (e.g., at the mid-10^5 cm^{-3} level or higher) is required to initiate nucleation under most of the atmospheric conditions, although NPF does not always take place even at high sulfuric acid conditions. In many studies, the reported sulfuric acid concentrations were not measured but have been calculated from SO_2 and radiation using the statistical correlation of Mikkonen et al. (2011), for example. While J has a strong correlation with sulfuric acid, it usually has a much weaker correlation (sometimes even an anticorrelation) with base compounds, such as ammonia and amines (Erupe et al., 2010; Kürten, Bergen, et al., 2016; McMurry et al., 2005), despite evidence from laboratory studies that base compounds are enhancing J (Almeida et al., 2013; Jen et al., 2014; Kirkby et al., 2011; Yu, McGraw, & Lee, 2012). Field observations also show an opposite diurnal cycle of NPF and HOMs (Bianchi et al., 2017; Kürten, Bianchi, et al., 2016; Rose et al., 2018; Yan et al., 2016), suggesting that the role of HOMs in NPF is not well-understood.

One of the most important future studies is to conduct long-term measurements of NPF at diverse locations to understand how different NPF processes dominate under different atmospheric conditions. The majority of measurement sites with at least 1 year time series of aerosol size distributions are located in Europe, while long-term data from Asia, North America, and the Southern hemisphere are very limited (Nieminen et al., 2018). For example, in North America, long-term observations NPF have been conducted at limited locations (Hallar et al., 2015; Kanawade et al., 2012; Sullivan et al., 2018). Simultaneous measurements of aerosol size distributions and chemical analysis of the precursors are especially scarce. Many of the existing time series are too short to study the seasonal variation and trends of NPF over long time periods. Several studies showed that dominant nucleation and growth mechanisms can vary even at the same site depending on the availability of precursor vapors (Bianchi et al., 2016; Hodshire et al., 2016), indicating that long-term observations are needed before drawing conclusions about the prevailing nucleation mechanisms.

4.1. Atmospheric Observations of HOMs and Their Implications on NPF

The longest semicontinuous HOM measurements from the boreal forest area were conducted with a nitrate-CIMS at Station for Measuring Forest Ecosystem – Atmosphere Relations SMEAR II station in Hyytiälä in the southern Finland (Bianchi et al., 2017; Yan et al., 2016). These measurements show that HOMs (containing up to 40 carbons) are relatively abundant, with maximum total HOM concentrations during the spring, with concentrations more than 1 order of magnitude larger than those of sulfuric acid. The concentrations of HOMs were usually higher during the night, whereas HOM-ONs were higher during the daytime. For example, HOM dimers show the opposite diurnal trend to sulfuric acid (i.e., a maximum at the night and a minimum during the day).

Furthermore, these measurements indicate that it is not possible to explain the measured GR with the measured sulfuric acid (10^6 cm^{-3}) and HOMs (10^7 cm^{-3}) in the atmosphere, since they together can explain only a small fraction of the measured GR (up to tens of nanometers per hour), even assuming that HOMs also condense on new particles with the collision-limited process (Bianchi et al., 2017; Yan et al., 2016). Hence, it is likely that a large fraction of oxidized organics (especially LVOCs and SVOCs) are not currently measured with the nitrate CI-APi-TOF, and alternative techniques are needed for their detection.

The nighttime composition of HOMs in the boreal forest is similar to the ozonolysis products of α-pinene found in chamber experiments (Bianchi et al., 2017; Yan et al., 2016). This reflects the fact that α-pinene from coniferous trees is the most abundant monoterpene in boreal forests (Rinne et al., 2000), although other VOCs, for example, Δ^3-carene and β-pinene, as well as sesquiterpenes, can also contribute to HOMs formation. Although nighttime NPF events are rare in boreal forests, HOM clusters may be responsible for frequent formation of larger ion clusters during the nighttime (Rose et al., 2018). In a rural site in central Germany, the measured HOMs were also more abundant during the nighttime, whereas NPF was observed during the day (Kürten, Bianchi, et al., 2016). The reason for the lack of nighttime NPF in these

environments is not known, but it is possible that there are not enough other low volatile vapors (e.g., HOM-ONs) available to grow the HOM clusters.

During the day, a large fraction of HOMs found in the boreal forest contains one or two nitrate groups (i.e., HOM-ONs; Bianchi et al., 2017; Yan et al., 2016). The role of HOM-ONs in particle formation is still under debate. Field studies show good correlation between some of the HOM-ONs and particle formation (Jokinen et al., 2017). Laboratory studies, on the other hand, indicate that HOM-ONs are in general more volatile and thus less likely to form particles than nonnitrate HOMs, at least on their own (Lehtipalo et al., 2018).

Atmospheric observations show sometimes distinctive shrinkage (as opposed to growth) events of newly formed particles, usually associated with high temperatures (Alonso-Blanco et al., 2017; Cusack et al., 2013; Salma et al., 2016; Skrabalova et al., 2015; Yao et al., 2010; Young et al., 2013; Zhang et al., 2016). These observations imply that SVOCs are also involved in the growth of new particles. However, apparent shrinkage of the mean diameter of a particle population can also be caused by spatial variations in vapor production and GR (Kivekäs et al., 2016).

4.2. Ion Nucleation

Observations of atmospheric ions and their connection to nucleation has been reviewed by Hirsikko et al. (2011). Air ions are ubiquitous in the lower troposphere (Horrack, Mime, et al., 1998; Horrack, Salm, et al., 1998). Prenucleated ion clusters have been measured, and the results in general show that negative clusters play more important roles in aerosol nucleation in lower troposphere than positive ions (Eisele, 1988; Eisele et al., 2006; Eisele & Hanson, 2000). Large ions, both positive and negative, containing inorganic and organic components, were also observed in the upper troposphere (Eichkorn et al., 2002). S. H. Lee et al. (2003) showed that ion nucleation is important in the upper troposphere and lower stratosphere, while other studies have not seen such effect (Mirme et al., 2010).

Based on long-term measurements of ions made in the boreal forest in the southern Finland, Kulmala et al. (2013, 2000) concluded that the contribution from ions is negligible in the boreal forest. On the other hand, IMN modeling simulations showed that ion-ion recombination plays critical roles for aerosol nucleation in the boreal forest (Yu, 2011; Yu & Turco, 2008). Recent observations also suggest that ion nucleation may play a role in the formation of large clusters during the nighttime in the boreal forest (Junninen et al., 2008; Rose et al., 2018). A multisite study has shown that boundary layer ion formation rates are in general 1–30% of the corresponding total J depending on the location (Manninen et al., 2010). Ion nucleation is generally more important in the chemical systems when there are no other species available to stabilize the forming clusters (Wagner et al., 2017). The role of ions in different nucleation schemes and under different atmospheric conditions remains to be quantified.

4.3. NPF in Extremely Polluted Conditions

There is a conventional wisdom that NPF should not occur under high concentrations of preexisting aerosols, because nucleation precursors condense on preexisting particles and small particles are scavenged by high concentrations of large particles. Thus, the large condensation sink (CS, which is proportional to the total surface area of preexisting aerosols) should suppress NPF. However, very frequent NPF has been observed in extremely polluted megacities, as also summarized by Chu et al. (2019). Figure 2 shows NPF takes place under high CS conditions in urban Shanghai (Yao et al., 2018). In particular, the GR is also higher than those found in many places around the world with much lower CS (Table 1). In this section, we discuss the primary quantities used to characterize atmospheric NPF events, occurrence frequency, J, GR, new particle number concentration (N), and CS, in polluted environments.

The observed CS is on the order of 0.01 to 0.1 s^{-1} in polluted urban locations (Table 1), which are 1 or 2 orders of magnitude higher than those in rural or remote environments (Kulmala, Vehkamäki, et al., 2005). Nucleation measurements at ~1 nm show that J values in three megacities, Mexico City, Nanjing, and Shanghai, for example, are 2 orders of magnitude higher than those observed in the boreal forest (Kulmala et al., 2013). A global comparison of sub-3-nm particle number concentrations (N_{sub-3}) further indicates that N_{sub-3} in polluted megacities could be up to 2 orders of magnitude higher than in clean and moderately polluted environments (Kontkanen et al., 2017). When particles grow to 3–15 nm, the J values decay to a few to tens of new particles per cubic centimeter per second but are still higher than, or at least comparable to, those in rural and remote environments (Kulmala, Vehkamäki, et al., 2004). Particle GR in

Table 1
A Summary of Formation Rate (J), Growth Rate (GR) in >3-nm Sizes, and Condensation Sink (CS) Reported in Highly Polluted Environments

Location	J (cm^{-3} s^{-1})		GR in >3-nm sizes (nm/hr)	CS (s^{-1})	Reference
Mexico City, Mexico	J_1	9–5,000	0.5–9.0		M. Chen et al. (2012) and Dunn et al. (2004)
Nanjing, China	$J_{1.4}$	92–2,500	5.5–10.1	0.016–0.033	H. Yu et al. (2016)
Shanghai, China	$J_{1.34}$	112–271	4.5–38.3	0.03–0.10	Xiao et al. (2015)
Beijing, China	J_3	3.3–81.4	0.1–11.2	0.006–0.06	Wu et al. (2007)
Beijing, China	J_3	2.2–34.5	2.5–15.3	0.027 ± 0.021	Z. B. Wang et al. (2013)
New Delhi, India	J_3	3.3–13.9	11.6–18.1	0.05–0.34	Mönkkönen et al. (2005)
Pune, India	J_5	3.5–13.9	6.1–7.6	0.0162 ± 0.007	Kamra et al. (2015) and Kanawade et al. (2014)
Kanpur, India	J_5	0.4–3.2	5.2–13.3	0.0333 ± 0.008	Kanawade et al. (2014)
Nanjing, China	J_6	0.24–10.9	3.6–23	0.007–0.068	Qi et al. (2015)
Taichang (YRD region), China	J_{10}		6.4 ± 1.6	0.024–0.082	Gao et al. (2009)
PRD region, China	J_{15}	2.4–4.0	4.0–22.7	0.023–0.046	D. L. Yue et al. (2013)
Wuxi (YRD region), China	J_{15}		6.2–13.3	0.009–0.028	Peng et al. (2014)

urban locations in general tends to be higher than in rural or remote environments (Kulmala et al., 2004) but are usually still within the same order of magnitude.

Long-term measurements of NPF showed diverse temporal variations in different urban locations. The annual averaged frequencies of NPF in Beijing (40%) and Nanjing (44%) of Northern China (Qi et al., 2015; Wu et al., 2008) are clearly higher than in the Pearl River Delta region in the southern China (13%; Wang, Hu, Mogensen, et al., 2013) and Pune (26%) and Kanpur (14%) in India (Yue et al., 2015). Distinct seasonal variations of NPF events were also observed. For example, NPF in Beijing was characterized by high frequencies during the spring and winter than the summer. In comparison to the boreal forest, the Helsinki urban atmosphere also showed a less distinctive seasonal difference in N_{sub-3} between winter and summer (Kontkanen et al., 2017). The fundamental reasons for these differences imply the balance between factors that contribute to or suppress NPF and growth.

Unlike regional NPF in a large-scale uniform air mass, NPF in polluted urban areas is dominated and complicated by local sources like industrial plumes (Sarnela et al., 2015) and vehicle emissions (Alanen et al., 2015; Karjalainen et al., 2016). Spatial variations of nucleation and growth in inhomogeneous air masses thus lead to different evolution patterns of the particle size distribution observed at a fixed monitoring location (Kivekäs et al., 2016). Dai et al. (2017) observed that NPF events in Nanjing were sometimes connected to AVOC plumes emitted from nearby industrial areas. In those local NPF events, nucleation was estimated to occur within a distance of ~50 km upwind of the observation site. As a result, further growth beyond 20 nm was not followed at their measurement site.

Correlation analysis between NPF and chemical and environmental variables provides indirect evidence of the NPF mechanisms. There is strong evidence that NPF events at polluted urban locations are linked to sulfuric acid (Kanawade et al., 2014; Kontkanen et al., 2017; Mönkkönen et al., 2005; Nie et al., 2014; Shen et al., 2016; Xiao et al., 2015). Particle formation rates have been shown to be positively correlated with concentrations of ammonia (Xiao et al., 2015) in Shanghai and amines (Yao et al., 2016) (e.g., Figure 2). Nie et al. (2014) and Xie et al. (2015) also proposed that heterogeneous photochemistry between SO_2 and NO_2 induced by dust storm and biomass burning particles may produce sulfate and nitrous acid (HONO). HONO produces additional gaseous oxidant OH upon photodissociation, which enhances the atmospheric oxidation capacity and the probability of NPF. This heterogenous reaction mechanisms need to be verified in future laboratory studies.

The relationship of NPF to temperature is not consistent at different locations. Positive correlations between NPF and temperature were observed in Shanghai (Xiao et al., 2015), Kanpur (Kanawade et al., 2014), and Singapore (Betha et al., 2013), whereas negative correlations were observed in Nanjing (Yu et al., 2016). On the basis of the correlation analysis, several studies modeled the power law dependence of measured formation rates on measured/modeled sulfuric acid and organic vapors under polluted atmospheric conditions (Beijing and Nanjing; X. Huang et al., 2016; J. Wang & Wexler, 2013; Z. B. Wang et al., 2015; Z. B. Wang et al., 2011). By doing so, different nucleation schemes of monomolecular nucleation, heteromolecular nucleation, or a combination of both were proposed.

Resolving high J and GR under high CS conditions will require coordinated field studies that employ a large set of chemical and aerosol instruments at several different urban sites, along with comprehensive measurements of trace gas species and meteorological parameters. Finding out the potential chemical precursors that are responsible for NPF in urban areas, as well as the synergetic effects between inorganic and organic precursors and between BVOCs and AVOCs, under different oxidation conditions will help to understand the NPF mechanisms in megacities.

4.4. Coagulation Removal Versus Fast Growth in Polluted Areas

It would be tempting to argue that NPF frequently observed in highly polluted environments is due to the abundant supply of low volatile vapors that may outcompete coagulation scavenging by preexisting aerosol particles. However, calculations using the Kerminen-Kulmala equation (Kerminen & Kulmala, 2002) still yield extremely low survival probabilities of 10^{-4}–10^{-17} for 1.5 nm clusters to grow to 3 nm, under the conditions with high CS/GR ratios typically found in polluted environments (Kulmala et al., 2017). This indicates that new particle formation should not occur in highly polluted environments, based on the current knowledge of NPF.

From the perspective of the survival probability and the aerosol population dynamic balance, respectively, Kulmala et al. (2017) and Dai et al. (2017) suggested that the detectable NPF under the highly polluted conditions can be explained only if the coagulation sticking probability (β) of molecular clusters with preexisting particles is considerably less than 1, or the GR of 1- to 3-nm particles is much faster than typical values provided by the current GR measurement methods or a suitable combination of both. As demonstrated in Figure 5 (right panel), the estimated GR from the aerosol dynamic balance drastically varies with different coagulation sticking probabilities ranging from 0.1 to 1. The evidence from the survival probability and the aerosol population dynamic balance thus strongly imply that CS/GR values derived from the conventional estimation methods (Kulmala et al., 2012) may be overestimated by a factor of ~20 for sub-3-nm particles in the highly polluted urban atmosphere of Nanjing (Kulmala et al., 2017; Yu et al., 2016).

Theoretical and experimental data of coagulation probabilities between clusters and particles are not available at present. The coagulation sticking probabilities may be considerably less than 1, considering the fact that high kinetic energy of small particles of sub-10 nm can cause the particle "bounce-off" (Hollander et al., 1995; Narsimhan & Ruckenstein, 1985). In other words, it is possible that small clusters, at least in extremely polluted environments where most of preexisting particles are freshly emitted from primary sources, can overcome the heterogeneous nucleation barrier before being incorporated into the bulk particulate phase (Kulmala et al., 2017).

The high "apparent" CS/GR ratios found in highly polluted environments can also be due to the underestimation of GR in the sub-3-nm size range. If clusters are charged, condensation flux of vapors onto charged clusters is expected to be enhanced. On the other hand, Lehtipalo et al. (2016) found no enhancement effects of charges on GR, in the presence of amines. Also, multiphase chemical processes, such as solvation effects (Kulmala et al., 2004), surface adsorption (J. Wang & Wexler, 2013), and heterogeneous chemical reactions (Zhang & Wexler, 2002) between organic activating vapors and clusters (Zhang & Wexler, 2002), could help to overcome the Kelvin effect and facilitate the faster condensation of activating vapors onto 1 to 3 nm particles. In addition, the contribution of self-coagulation of clusters to the growth under highly polluted conditions may need to be reevaluated, although calculated cluster-cluster collision rates are usually negligible based on the current knowledge (Yao et al., 2018).

4.5. NPF in Coastal Regions

NPF has been long observed in the coastal regions of Ireland, Scotland, France, Australia, Antarctica and China (Bigg & Turvey, 1978; Flanagan et al., 2005; Huang et al., 2010; O'Dowd et al., 2002; Saiz-Lopez et al., 2012; Wiedensohler et al., 1996; Y. Hu et al., 2019). The North Atlantic Aerosols and Marine Ecosystems Study NAAMES campaign has elucidated the contribution of NPF involving sulfur to CCN concentrations in the marine atmosphere (Sanchez et al., 2018). In addition to sulfuric acid, methanesulfonic acid (MSA) formed from oxidation of dimethyl sulfide (DMS) may contribute to NPF, as shown by CNT calculations (Kreidenweis et al., 1989), quantum chemical calculations (Bork et al., 2014; Wen et al., 2018), and flow tube experiments (Chen et al., 2015; Dawson et al., 2012; Ezell et al., 2014). Although MSA concentrations are comparable to sulfuric acid in the marine boundary layer, laboratory measurements showed that J of
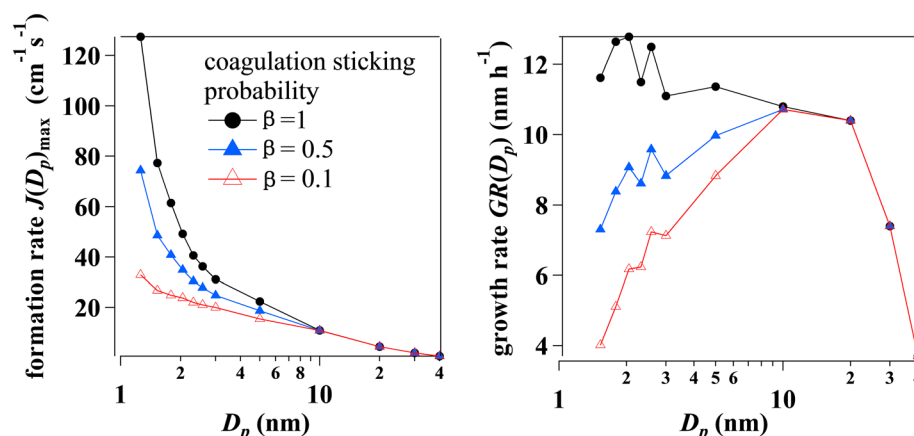
Figure 5. Size-dependent formation rate and growth rate estimated from aerosol dynamic balance in a NPF event on 6 November 2015 in Nanjing for an assumed coagulation sticking probability, β, of 0.1, 0.5, and 1.0 for sub-10-nm particles. Adapted from Dai et al. (2017).

binary nucleation of MSA and water are relatively small (Wyslouzil et al., 1991). Recently, H. Chen et al. (2016) showed that MSA can form particles with ammonia and determined their cluster formation free energies. Dawson et al. (2012) developed a kinetic model for the formation of particles from MSA and amines employing quantum chemical calculations combined with flow tube experiments.

Iodine concentrations in aerosols are considerably enriched over the seawater composition, as indicated by the measured I/Cl and I/Na ratios (Baker et al., 2000). This enrichment suggests there is a route for iodine species in the seawater to be converted into particles in the ambient air. Iodinated hydrocarbons (e.g., CH_3I, CH_2ICl, CH_2IBr, and CH_2I_2) and molecular iodine (I_2) emitted from macroalgae, microalgae, and ocean surface iodide activation are the known sources of volatile iodine compounds. The total oceanic emission of iodine species is estimated to be ~1 Tg I/year (Simpson et al., 2015). Strong correlations of daytime tidal cycles, iodocarbons, iodine monoxide (IO), and ultrafine particle bursts provide the strong evidence for the role for iodine compounds for NPF in the coastal atmosphere (Commane et al., 2011; Furneaux et al., 2010; Huang et al., 2010; McFiggans et al., 2010; Saiz-Lopez et al., 2012; Simpson et al., 2015; Whitehead et al., 2009), although this does not exclude the potential participation of sulfuric acid in nucleation and condensational growth. Vaattovaara et al. (2006) observed that organics also contribute to the growth of freshly formed coastal (iodine) particles. Recently H. Yu et al. (2019) have shown that previously unidentified organic iodine compounds, such as iodate, aromatic iodinecompounds, iodoacetic acid or iodopropenoic acid, and iodide–organic adducts can also participate in NPF in an iodine hotspot at the east coast of China.

Based on the current knowledge, the simplified schematic of atmospheric iodine oxidation and nucleation processes can be described as the following: Volatile iodocarbons and molecular iodine are photolyzed to produce I atoms. I radicals are relatively less active toward VOCs, and hence, their main sink is the reaction with O_3 (which produces IO and IO_2 radicals). Self-recombination of IO and IO_2 produces higher oxides, I_2O_{2-5}. Transmission electron microscopy and mass spectrometry analysis indicated that the probable solid nucleating particles are composed of iodine oxides, for example, IO, IO_2, or I_2O_{2-5} (Gómez Martín et al., 2013; Jimenez et al., 2003; O'Dowd et al., 2002; Saunders et al., 2010; Saunders & Plane, 2006).

Iodine oxoacids HIO_x (X = 1–3) also form from IO/IO_2 reactions with HO_x (OH + HO_2) or hydration of I_2O_{2-5}. Sipilä et al. (2016) provided the field observation evidence in an Atlantic coastal site in Ireland that iodine cluster formation primarily proceeds by a sequential addition of HIO_3, followed by the subsequent dehydration which forms I_2O_5, using CI-APi-TOF. The measured O:I ratio of 2.4 suggests a less pronounced addition of HIO, HIO_2, and/or I_2O_{2-5}. Thus, there is most likely a humidity-dependent balance between clusters contacting HIO_3 and I_2O_{2-5} in the atmosphere. Quantum chemical calculations may help to further facilitate the development of full kinetic models for iodine nucleation (Khanniche et al., 2016).

It is also possible that NO_x and VOCs, which control OH and ozone formation in the atmosphere, can also affect formation of iodine oxides and iodine oxoacids in polluted coastal regions (Mahajan et al., 2009). The

roles of sulfur species (MSA and sulfuric acid) versus iodine species in marine and coastal, as well as in Arctic and Antarctic regions, are not yet well quantified. As in many parts of the world including China, emissions of SO_2 are expected to decrease; the relative importance of sulfuric acid versus iodide oxidants in NPF will become increasingly more relevant in the future coastal atmosphere.

4.6. NPF in Polar Regions

There has been increasing attention on NPF in the Arctic region (Abbatt et al., 2019; Asmi et al., 2016; Croft, Martin, et al., 2016; Dall'Osto et al., 2017; Freud et al., 2017; Heintzenberg et al., 2017; Tunved et al., 2013). Freud et al. (2017) compared long-term measurements of particle size distributions taken at five stations (Alert, Tiksi, Barrow, Nord, and Zeppelin) in the Arctic region and showed that the seasonal variation of NPF is quite uniform across the whole Arctic. NPF events seem to be restricted to summer months (mainly April–September) during the daytime, despite the presence of sunlight also during the summer nights. At the Siberian station in Tiksi, the highest formation rates were observed in spring, while the highest *GR* occurred later in summer (Asmi et al., 2016). Based on the comprehensive observations in the Canadian Arctic, Abbatt et al. (2019) concluded that NPF might be more favorable in the Arctic marine and coastal environments compared to the lower latitude marine atmospheres due to low *CS*, low temperatures, and longer sun exposure hours.

Particle size distributions have been measured at several locations in Antarctica, but only few extended to the sizes below 10 nm. NPF has been observed at the Aboa station, ~130 km from the coastline, with ~20% NPF frequency during the austral summer (Asmi et al., 2010; Jokinen et al., 2018; Koponen et al., 2003; Kyrö et al., 2013; Virkkula et al., 2007). NPF has been observed also at continental Antarctica at Concordia station at Dome C on 5–25% of days depending on the season, with the highest occurrence frequency during the summer (Järvinen et al., 2013). Interestingly, NPF occurred sometimes also in the absence of sunlight. However, the formation rates (J_{10}) at Dome C were about 10 times lower (median 0.002 cm^{-3} s^{-1}) than those measured at Aboa (Järvinen et al., 2013). The median *GR* of nucleation mode particles were between 1 and 5 nm/hr at both locations.

Sulfur compounds (Giamarelou et al., 2016; Ito, 1993) from marine DMS emissions (Burkart et al., 2017; Chang et al., 2011) have been proposed as nucleation precursors in polar regions, with possible influences of ammonia from seabird colonies (Croft, Wentworth, et al., 2016) and iodine compounds (Allan et al., 2015; Sipilä et al., 2016). The first measurements of molecular composition of nucleating clusters in the coastal Antarctica indicated that the prevailing mechanism is ion nucleation of sulfuric acid and ammonia (Jokinen et al., 2018). Future studies are required to determine the chemical composition of the vapors participating in NPF across the polar regions with comprehensive chemical measurements.

4.7. NPF at High Elevation

There are several potential underlying principles that allow nucleation to occur more frequently at high elevations. First, high elevations have lower temperatures and lower *CS* than at lower elevations. Also, high elevations have a favorable thermodynamic condition for NPF due to turbulence, atmospheric waves, storms, and other air mixing processes (Easter & Peters, 1994; Khosrawi & Konopka, 2003; Nilsson et al., 2000; Nilsson & Kulmala, 1998; Nyeki et al., 1999; Young et al., 2007). Turbulence on various scales including boundary layer turbulence (Nilsson et al., 2001), atmospheric waves (Nilsson et al., 2000), storms (Clement et al., 2002), and mixing across temperature and humidity gradients (Kulmala et al., 1998) may promote nucleation. These mixing conditions can create higher supersaturation ratios of nucleation precursors by mixing humid air with colder temperatures. Convection can also bring nucleation precursors from the ground level to the free troposphere where temperatures and *CS* are much lower (Benson et al., 2008; Lee et al., 2003). Ion production rates by cosmic rays are strongest at the altitudes around 10 km (Reiter, 1992), so ion nucleation can also take place at high elevation. Measurements above the boundary layer are needed to evaluate the effects of NPF on the global climate. Table 2 summarizes the NPF measurements made at mountain sites over the world, including NPF frequencies, particle formation rate, and *GR*.

Using two different high elevation sites, the Puy de Dôme research station (1,465 m above sea level [a.s.l.]) and the Opme Station (660 m a.s.l.) in central France, Boulon et al. (2011) illustrated the importance of vertical extent for nucleation events. The NPF frequency was greater (36% frequency in occurrence) at the

Table 2

Summary of the Observed NPF Frequency, Particle Formation and Growth Rates at High-Elevation Sites

Location/altitude (a.s.l.)	Study period	Frequency of NPF	Formation rate (cm^{-3} s^{-1})	Growth rate (nm/hr)	References
Mt. Yulong (3,410 m), China	May to Apr 2015	14%	1.33	3.22	Shang et al. (2018)
Mt. Tai (1,534 m), China	July to Dec 2014 June to Aug 2015	40%, 164 days	0.82–25	0.58–7.76	Lv et al. (2018)
	July 2010 to Feb 2012	32%, in the spring days	4	6.1	Shen et al. (2016)
Mt. Heng (1,269 m), China	Mar to May 2009		0.15–0.45	4.3–7.7	Nie et al. (2014)
Mount Waliguan (3,816 m), China	Sep 2005 to May 2007				Kivekas et al. (2009)
Mt. Werner (3,220 m), Colorado, USA	2001 to 2009	52%, 474 days	0.39	7.5	Hallar et al. (2016)
Izaña Mountain (2,373 m), North Atlantic	June 2008 to June 2012	50–60%	0.46	0.43	García et al. (2014)
Mukteshwar, Himalaya foothills (2,180 m), India	2005 to 2010	64% during the dry season	0.44	2.47	Neitola et al. (2011)
Mahabaleshwar (1,351 m), India	Nov 2016 and Feb 2017	47%, 115 days	2.8	2.6	Singla et al. (2018)
Mt. Lemmon, Arizona (2,790 m), USA	Feb to May 2002	112 days		12	Shaw (2007)
Puy de Dôme (1,450 m), Central France	Feb 2007 to June 2010	36%, 157 days	1.38	6.20	Boulon et al. (2011)
Jungfraujoch (3,580 m), Switzerland	Apr 2008 to Apr 2009	18%, 309 days	0.9	6.0	Bianchi et al. (2016) and Boulon et al. (2010)
Chacaltaya (5,240 m), Bolivia	Jan to Dec 2012	64% and maximum up to 100% in dry season	Dry season: 1.90 Wet season: 1.02	3.64–7.62	Rose et al. (2015)
Mt. Norikura (2,770 m), Japan	Sep 2001 and Aug to Sep 2002	17%, 23 days		2.85	Nishita et al. (2008)
Himalayas (5,079 m), Nepal	16 months	>35% during 511 days	0.35	1.8	Venzac et al. (2008)
Maïdo Observatory, Indian Ocean (2,150 m)	May 2014 to December 2015	65%	0.9–0.5	15.16	Foucart et al. (2018)

higher elevation site compared to the lower elevation site (21%). Frequent NPF events occurred at Mt. Werner (3,220 m a.s.l.) in Colorado, during 52% of the 474 measurement days from 2001 to 2009 (Hallar et al., 2011). Venzac et al. (2008) have reported a NPF frequency of higher than 35% during 511 days including monsoon and post monsoon seasons at the Himalayas in Nepal (5,079 m a.s.l.). They showed the high NPF frequency of 64% during the dry season (May–October). Several studies on NPF has been made in high mountains in China, such as Mt. Tai (Lv et al., 2018), Mt. Heng (Nie et al., 2014), Mount Waliguan (Kivekas et al., 2009), and Mt. Yulong (Shang et al., 2018). Lv et al. (2018) conducted observations during the different seasons in 2004 and 2005 at the summit of Mt. Tai (1,534 m a.s.l.) in the eastern China. Sixty-six NPF events were identified during 164 days of measurements with an occurrence frequency of ~40%.

These cited studies have only measured aerosol size distributions so far, and measurements of ions and the chemical analysis of nucleation precursors at high-elevation sites are limited. At Jungfraujoch, Switzerland (3,580 m a.s.l.), NPF was found on 18% of the 309 measurement days (Boulon et al., 2010). Recently, using a series of chemical analysis at the same site, Bianchi et al. (2016) showed that some of the NPF events were associated with high concentrations of HOMs, unlikely chemical species expected in the free troposphere. NPF occurred mainly through condensation of HOMs, in addition to taking place through sulfuric acid-ammonia nucleation. Neutral nucleation was more than 10 times faster than ion nucleation, and the *GR* were size-dependent. NPF occurred during a time window of 1 to 2 days after the contact with air masses originated from the planetary boundary layer. While ion nucleation can be expected at high elevation, at Puy de Dome, Rose et al. (2018) observed that neutral clusters dominate during NPF. Continuous measurements of ions at high-elevation sites are required to elucidate the role of ions at high elevation. Simultaneous lidar measurements of the boundary layer dynamics are also required to understand the effects of convection and air mixing on NPF at high-elevations sites. How long-range transported plumes, including biomass burning plumes, affect NPF is another important scientific question.

4.8. Isoprene Suppression of NPF

Absence of NPF in Amazon forests is one of the most striking features in NPF, especially in contrast to the prevalent NPF events observed in many parts of the world, despite the high concentrations of monoterpenes which chamber measurements suggested should oxidize and nucleate (Gordon et al., 2016; Kirkby et al., 2016). In the last decades, measurements of submicrometer aerosol size distributions have been undertaken in successive intensive field campaigns in the Amazon rainforest (Martin et al., 2016; Rizzo et al., 2018; J. Wang et al., 2016). However, these studies consistently demonstrated the absence of NPF in Amazon, either at forest sites (Rizzo et al., 2010; Zhou et al., 2002) or at the sites influenced by biomass burning emissions (Rissler et al., 2004).

Absence of NPF has been also reported from observations in other forests in the United States, with dominant isoprene emissions, for example, in a rural forest in Alabama (Lee et al., 2016), Michigan (Kanawade et al., 2011), Whiteface Mountain (Bae et al., 2010), Pinnacle State Park (Bae et al., 2010), Duke forests (Pillai et al., 2013), and the high-elevation mountain-top site in Steamboat Spring in Colorado (Hallar et al., 2015; Yu et al., 2015). However, most of these studies did not investigate the mechanisms behind the absence of NPF.

One explanation for the absence of NPF is that isoprene can suppress NPF, as suggested by biogenic plant experiments (Kiendler-Scharr et al., 2009; McFiggans et al., 2019). The plant chamber studies showed that isoprene inhibits the OH production in the chamber to suppress NPF (Kiendler-Scharr et al., 2009). However, in real forest environments, OH is not depleted and often regenerated. For example, the measured OH in Michigan and Amazon forests (1×10^6 cm^{-3} noontime peak) were relatively high (Di Carlo et al., 2004; Kanawade et al., 2011; Martinez et al., 2010), consistent with OH measurements in other forests (Hofzumahaus et al., 2009; Lelieveld et al., 2008; Paulot et al., 2009; Peeters et al., 2009). Currently, it is not clear what chemical mechanisms are behind the suppression of NPF in isoprene-dominated forests. S. H. Lee et al. (2016) showed that in the case of the Alabama forest, there were sufficient precursors such as sulfuric acid, ammonia, amines, and relatively low *CS*, all comparable to the conditions that should allow NPF to take place. Furthermore, they found that there was a clear formation of sub-3-nm particles, which were strongly correlated to sulfuric acid concentrations, but these clusters did not grow further. From these results, they hypothesized that isoprene oxidation products are too volatile compared with oxidation products of monoterpenes and compete with monoterpene oxidation products for interactions with sulfuric acid clusters.

In the case of the Amazon region, there may be other reasons that can also explain the absence of NPF. First, the level of gaseous nucleation precursors (e.g., sulfuric acid and ammonia) in the Amazon under pristine conditions may be too low to initiate nucleation (Andreae et al., 1990; Trebs et al., 2004). Pöhlker et al. (2012) observed the pronounced biogenic potassium content in Amazon organic aerosols and suggested that NPF may be suppressed by the presence of biogenic particles acting as seeds for condensation of aerosol precursors. J. Wang et al. (2016) have shown that NPF takes place in the cold free troposphere, and these nuclei mode particles are then transported downward by convective movements and become a source of residual concentrations of nuclei mode particles observed at the ground level in Amazon, but these particles still do not grow to show the typical "banana" plots.

4.9. Nighttime Nucleation

Because sulfuric acid is the key nucleation precursor and its production is generally correlated with solar flux, nucleation should occur during the daytime. However, there is evidence from aircraft measurements over a large range of latitudes and altitudes that nucleation also takes place during the night in the free troposphere (Hermann et al., 2003; Lee et al., 2008). Observations at the ground level have shown some nighttime particle formation events at different locations (Buenrostro Mazon, 2016; Duplissy et al., 2008; Junninen et al., 2008; Kalivitis et al., 2012; Lehtipalo et al., 2011; Liu et al., 2014; Man et al., 2015; Ortega et al., 2009; Ortega et al., 2012; Pushpawela et al., 2018; Rose et al., 2018; Russell et al., 2007; Salimi et al., 2017; Suni et al., 2008; Svenningsson et al., 2008; Wiedensohler et al., 1997; Zhu et al., 2014). Nighttime nucleation is characterized by enhanced nuclei mode particle concentrations, but they usually do not show the typical "banana" shaped growth as seen during the daytime NPF events.

The mechanisms behind nighttime nucleation are very uncertain. OH radicals can be produced from ozonolysis of monoterpenes even at night (Donahue et al., 1998; Paulson et al., 1997; Paulson et al., 1999), which in turn react with SO_2 to produce sulfuric acid and new particles. Mauldin et al. (2012) suggested that stabilized Criegee intermediate oxidants, produced from monoterpene ozonolysis, can react with SO_2 to produce sulfuric acid. On the other hand, long-term measurements of sulfuric acid in the boreal forest consistently show distinctive diurnal variations with very low nighttime concentrations of sulfuric acid, even with abundant monoterpenes (Petäjä et al., 2009). At present, it is not clear how these reaction pathways involving stabilized Criegee intermediate can affect atmospheric sulfuric acid formation (Novelli et al., 2017) and NPF. Recently, Kirkby et al. (2016) have shown that biogenic nucleation can occur assisted by ions even in the absence of sulfuric acid. It is possible this nucleation process can contribute to the nighttime NPF observed in some forests (Rose et al., 2018).

One feature of the nighttime NPF is that it often takes place after the sunset when air masses are influenced by the transported SO_2 plumes. For example, intense bursts of nuclei mode particles were observed in rural forests in Michigan and Alabama with elevated SO_2 concentrations (at ppbv), while NPF did not occur even during the daytime under low or moderate SO_2 concentrations (at sub-ppbv; Kanawade et al., 2011; Lee et al., 2016). It was hypothesized that some heterogeneous oxidation processes can produce sulfuric acid on small particles (Lee et al., 2008), similarly to those found on cloud and fog droplets (Finlayson-Pitts & Pitts, 2000; Seinfeld & Pandis, 2016) and acidic aerosol particles (Hung & Hoffmann, 2015; Stangl et al., 2019).

5. Parameterization of NPF in Atmospheric Models

In order to represent NPF in climate, air quality, and numerical weather prediction models, expressions for the NPF rate as a function of relevant vapor concentrations, temperature, RH, and atmospheric ion concentrations are needed. When implemented in atmospheric models, the influence of these parameterizations on atmospheric particle concentrations and radiative forcing of climate is strongly dependent not only on the details of the parameterization but also on the rest of the model. Therefore, it is important for parameterizations to be based on experimental or theoretical results rather than tuned to reproduce observed particle number concentrations, to avoid obtaining realistic particle concentrations by compensating for other model biases.

In this section, we review the current status of parameterizations of NPF and their implications when implemented in atmospheric models. We focus on homogeneous nucleation theories including ion nucleation and on the troposphere rather than the stratosphere. However, other processes, such as heterogeneous nucleation (Tauber et al., 2018) may be also important. Recent theoretical work on cluster-substrate interactions may result in new parameterizations of heterogeneous nucleation appropriate for atmospheric models in future (McGraw et al., 2018). Appendix B shows several NPF parameterizations retried from the CLOUD experiments.

5.1. CNT for Binary Sulfuric Acid-Water Nucleation

NPF in the majority of atmospheric models is parameterized from CNT (Becker & Döring, 1935; Flood, 1934; Volmer & Weber, 1926), as described in section 1. Maattanen et al. (2018) made various improvements on the (Vehkamäki et al., 2002) CNT parameterization, based on theoretical developments by Merikanto et al. (2016) and hydrate distributions from Kurten et al. (2007). The theoretical developments of Merikanto et al. (2016) were carefully tested against measurements at the CLOUD chamber (Duplissy et al., 2016). Reasonably good agreement is found, although the theory predicts higher J than is observed at the lowest temperatures, sometimes exceeding the experimental data by more than a factor of 10. At these temperatures, however, uncertainties in the measurement of the sulfuric acid concentration limit the accuracy of the comparison, while at high temperatures, the contamination of the chamber by ammonia is more important.

Alternative formulations of binary NPF, such as that of F. Q. Yu and Turco (1998), further improved on by F. Q. Yu (2005), are now being built on to develop parameterizations for more complex systems. F. Q. Yu (2005) employs a quasi-unary assumption, which assumes that sulfuric acid and water are always in equilibrium, so nucleation can be treated as a single-component process.

This simplification allows the pseudosteady-state approximation used in CNT to be avoided. Then, as done by Kazil and Lovejoy (2007), laboratory measurements of Hanson and Lovejoy (2006) replace the bulk surface tension, avoiding another key problem with older CNT.

5.2. Ion Nucleation

At slightly warmer temperatures and lower altitudes, evaporation rates of neutral clusters become larger, and ions may play a more important role in stabilizing them. Inorganic ion nucleation of sulfuric acid has been considered in CNT (Chan & Mohnen, 1980; Seinfeld & Pandis, 2016). Laboratory measurements of the thermodynamics of charged clusters (Curtius et al., 2001) permitted the development of the "kinetic model for Sulfuric-acid-water Nucleation" (SAWNUC; Lovejoy et al., 2004). The data have also been used by F. Q. Yu (2007) and Kazil and Lovejoy (2007) in kinetic models for ion nucleation, and these are extended to the ternary system by F. Yu et al. (2018). Pure CNT has also been applied by G. K. Yue and Chan (1979) and is still actively studied (Laakso et al., 2002; Maattanen et al., 2018; Merikanto et al., 2016). Future research on critical clusters in ion nucleation may benefit from recently developed tools such as the identity of McGraw et al. (2018) in CNT. Similarly to the neutral case, the ion nucleation rates from the SAWNUC and CNT models have since been compared to data from the CLOUD chamber by Ehrhart et al. (2016) and Duplissy et al. (2016), respectively. Both SAWNUC and CNT models agree reasonably well with the data, although the SAWNUC model was slightly modified compared to its original formulation (Ehrhart et al., 2016). Approximately, the J increases linearly with the ion concentration in typical conditions. Discrepancies between the model and the measurements are as likely due to systematic measurement uncertainties as to problems with the model.

Pure biogenic ion nucleation was parameterized by Kirkby et al. (2016) based on CLOUD measurements (together with neutral nucleation, but the neutral rate is very low in atmospheric conditions). The community awaits further research on the temperature dependence of this ion nucleation rate, as well as whether or not the rate varies linearly with the ion concentration as in the inorganic case.

5.3. Ternary Nucleation Systems

Nucleation involving a third species as ammonia was considered in CNT by Coffman and Hegg (1995), Korhonen et al. (1999), and Napari et al. (2002). However, early parameterizations produce unrealistically high particle number concentrations in atmospheric models and need to be scaled down by a factor 10^6 to yield better agreement with atmospheric observations (Jung et al., 2010). The temperature dependence of the recent CNT parameterization is also rather weak compared to CLOUD chamber measurements (Dunne et al., 2016).

Recent work has focused on addressing these shortcomings. The ACDC model (McGrath et al., 2012) was used to model the role of ammonia in nucleation by Henschel et al. (2016) and Olenius et al. (2013). Their results were implemented as a look-up table (linked to the publication) in the PMCAMx-UF by Baranizadeh et al. (2016). ACDC calculations for the ternary ion nucleation system were published by Kürten, Bianchi, et al. (2016) but did not include the most updated hydration calculations of Henschel et al. (2016). Ion nucleation is unimportant at very low temperatures, but even at 250 K, ions can substantially enhance low J. A ternary ion nucleation kinetic model by F. Yu et al. (2018) incorporating quantum chemical calculations provides the RH dependence and critical sizes. In parallel, laboratory measurements are used by McMurry and colleagues with a different acid-base kinetic reaction model (Chen et al., 2012; Jen et al., 2014; McMurry & Li, 2017). This model is presented in a dimensionless framework (McMurry & Friedlander, 1979) and has the advantage of applying (with appropriately chosen evaporation rates) to both ammonia and amines. The evaporation rates of clusters are derived by fitting the model to the J measured in a flow tube reactor. Analytical expressions for the J as a function of these evaporation rates are presented and could be implemented in atmospheric models.

Acid-base nucleation can also involve a range of amines, which are stronger bases than ammonia and therefore more efficient stabilizers of clusters (Kurtén et al., 2008). Along with other kinetic nucleation models (Paasonen et al., 2012), the ACDC code has also been used for DMA (Almeida et al., 2013), other monoamines (Olenius et al., 2017), and diamines (Elm et al., 2017). Stable clusters involving both ammonia and amines may further accelerate amine-driven nucleation (C. Y. Wang et al., 2018).

5.4. "Activation" and "Kinetic" Nucleation Parameterizations

Parameterizations for boundary layer nucleation have been derived by fitting regression coefficients to J measured in the field. Linear dependences on sulfuric acid are explained by the *activation* of a number of preexisting clusters (Kulmala et al., 2006), while quadratic dependences are explained by *kinetic* nucleation of sulfuric acid molecules stabilized by bases or organics (Kuang et al., 2008; Sihto et al., 2006; Weber et al., 1998). Different field locations suggest different prefactors depending on the concentrations of other participating vapors. Above the top of the boundary layer, the parameterizations must then be shut off artificially in models to avoid producing too many particles. Proxies for the organic compounds that participate in NPF (Paasonen et al., 2010) are also included, and the resulting parameterizations are similar to those from some chamber studies using organic proxy vapors (Metzger et al., 2010; Riccobono et al., 2014). Ions have also been included in field-based parameterizations (Nieminen et al., 2011).

We note that the smallest size at which nucleation rates are usually measured in the field is 3 nm, and uncertainties are then introduced, either in assuming that the losses of particles smaller than 3 nm in the model are the same as those at the location of the measurement or in extrapolating back to the critical size and then forward again to correct for the difference in loss rates using formulations like that of Kerminen and Kulmala (2002). Further uncertainties are introduced by the lack of a temperature or base dependence the majority of in these parameterizations. F. Q. Yu et al. (2010) showed that unsurprisingly, activation-type parameterizations derived for the continental boundary layer, which had then been applied globally (e.g., Merikanto et al., 2009), significantly overpredict the particle number concentrations over tropical and subtropical oceans.

5.5. Nuclei Mode Aerosol Microphysics

The growth and losses of newly formed particles must be either represented explicitly or parameterized to determine their effects on atmospheric aerosols and climate. The most precise atmospheric model that explicitly represents the growth of nanoparticles is that of Jung et al. (2010), where 41 size sections are employed in the TwO-Moment Aerosol Sectional (TOMAS) algorithm. The Advance Particle Microphysics APM scheme of F. Yu and Luo (2009) has similar high-resolution nuclei mode microphysics, while Global Model of Aerosol Processes (GLOMAP)-bin, Community Aerosol and Radiation Model for Atmospheres CARMA (Toon et al., 1988), and the extended Weather Research and Forecasting-Model for Simulating Aerosol Interactions and Chemistry WRF-MOSAIC scheme (Matsui et al., 2011) use 20 size sections, and most other models consider only lognormal modes. S. H. Lee et al. (2013) considered the length of time steps required in microphysics models to capture the equilibrium of gaseous sulfuric acid concentrations with a changing CS via the pseudosteady-state approximation (Pierce & Adams, 2009b).

Parameterized approaches to microphysics, often used to extrapolate from field measurements to the atmosphere as described in the previous section, generally employ the equation of Kerminen and Kulmala (2002), preferably in its improved form (Lehtinen et al., 2007)

$$J_x = J_1 \cdot \exp\left(-\gamma \cdot d_1 \cdot \frac{CoagS(d_1)}{GR}\right) \text{with } \gamma = \frac{1}{m+1}\left[\left(\frac{d_x}{d_1}\right)^{m+1} - 1\right], \tag{8}$$

to extrapolate the formation rate at size d_1, J_1, to that at size d_x, J_x. In this equation m is a constant with value between -1 and -2, and $CoagS$ is the coagulation sink for particles, which is directly proportional to CS for vapors at fixed m and depends strongly on particle size. To a first approximation, the GR of the smallest particles is proportional to the concentration of condensable vapors. As this concentration is inversely proportional to the CS, this means the particle survival probability is exponentially dependent on the square of the sink (Westervelt et al., 2014). In areas where $CoagS$ is low and GR is slow, self-coagulation of newly formed particles can be important (Anttila et al., 2010). For certain combinations of J and $CoagS$, changes in vapor concentrations due to the diurnal cycle may lead to very large biases in the formation rates of particles parameterized in this way (Olenius & Riipinen, 2017).

Several approaches to treat the condensation of organic molecules onto nuclei mode particles in models have been applied, the most popular being the Volatility Basis Set (VBS) (Donahue et al., 2011) approach (Glotfelty et al., 2017; Matsui, 2017; Pierce et al., 2011), although other methods are in active use (Yu,

2011). The Kelvin effect is considered for each volatility bin. Tröstl et al. (2016) found a strong sensitivity of CCN concentrations to the very earliest stages of particle growth. Their study was updated by Stolzenburg, Fischer, et al. (2018), who studied the temperature dependence of the growth rates and were able to exploit more precise measurements using improved instrumentation.

5.6. Modeling NPF in the Boundary Layer and Free Troposphere

Nucleation parameterizations for the boundary layer based on chamber or field measurements have been implemented in several atmospheric models. The simulated seasonal cycle of particle number concentrations at surface sites can be reproduced reasonably well by adding ammonia-sulfuric acid ternary nucleation to organic-sulfuric acid nucleation (Metzger et al., 2010; Paasonen et al., 2010; Riccobono et al., 2014). The winter background appears to need ammonia, while the summer maximum can only be explained with organics (Dunne et al., 2016). Ground-based measurements in mixed deciduous forests often show low NPF frequencies during the summer (Chu et al., 2019; Erupe et al., 2010; Kanawade et al., 2012; Kerminen et al., 2018; Kulmala, Vehkamäki, et al., 2004), while models often predict frequent NPF during the summer (Yu et al., 2015). None of the organic parameterizations used in current models (except the pure biogenic parameterization of Gordon et al. (2016), which is relatively unimportant in the present day atmosphere) are based on explicitly measured molecules from biogenic sources. Therefore, an element of model tuning is introduced when these parameterizations are used with the molecules from BVOCs available in models. Furthermore, the temperature dependence in these parameterizations is still very uncertain (Gordon et al., 2016; Yu, Luo, et al., 2017; Zhu et al., 2019), and bases such as ammonia or amines may still be required in reality. Concentrations of sulfuric acid in the gas phase are also uncertain, as the production rate depends on photochemistry which is affected by local cloudiness (Pietikäinen et al., 2014). Also, temperature dependence of organic NPF is likely to be extremely important (Dunne et al., 2016; Yu, Luo, et al., 2017). These large uncertainties hinder simulations of both the present day and preindustrial atmospheres and translate to large uncertainties in aerosol radiative forcing (Gordon et al., 2016).

Atmospheric emissions and losses of amines are not well constrained so are difficult to include accurately in models, although several groups have implemented simplified treatments (Bergman et al., 2015; Dunne et al., 2016; Yu & Luo, 2014a). Dunne et al. (2016) suggested amines participate in the formation of around 5% of new particles within 500 m of the surface, but a larger contribution, as high as 30%, cannot be excluded.

The current state of our understanding of the different molecules that participate in NPF for altitudes below around 5 km is summarized in Figure 6 (Gordon et al., 2017). It shows that in the present-day lower atmosphere, almost all nucleation involves sulfuric acid but usually with ammonia and organic molecules, while in the preindustrial atmosphere, ammonia was much less likely to be involved.

At present, the role of ternary vapors and of ion nucleation at high altitude remains uncertain. F. Q. Yu et al. (2010) found ion nucleation is important in both the middle and upper troposphere, whereas Dunne et al. (2016) found ions are only important compared to binary and ternary neutral nucleation in the midtroposphere. Ekman et al. (2012) found that nuclei mode particle concentrations at these altitudes were underestimated in the tropics. Remote sensing data can also be used in the model evaluation. For example, English et al. (2011) evaluated a range of nucleation mechanisms in the stratosphere against Stratospheric Aerosol and Gas Experiment II SAGE II data. Future modeling studies will need to include the contribution of organic compounds to NPF in the free troposphere, following observations over the Amazon (Andreae et al., 2018) and cloud-resolving model simulations (Ekman et al., 2008; Murphy et al., 2015).

5.7. The Influence of NPF on Airborne Particulate Matter

Since early work by Jacobson (2001) and others, a large number of regional models have specifically considered NPF in the context of air pollution. Jung et al. (2010) and R. Zhang et al. (2010) determined the influence of a range of nucleation parameterizations on $PM_{2.5}$ over the United States in the intercomparison study. Gaydos et al. (2007) considered the contribution of nucleation to particle number concentrations in the eastern United States using the PMCAMx model with TOMAS microphysics and a spatial resolution of 36 km.

Other polluted regions have also been studied extensively. NPF events in Beijing were well reproduced by the Weather Research and Forecasting (WRF) model with additional size bins in the Model for Simulating Aerosol Interactions and Chemistry aerosol scheme by Matsui et al. (2011). Empirical
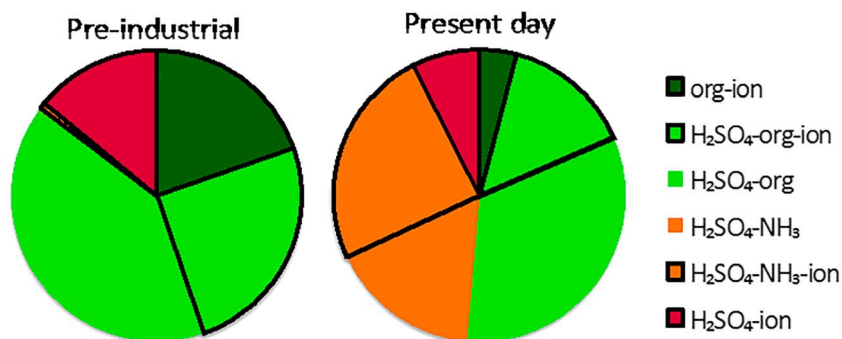
Figure 6. The relative contributions of nucleation processes involving both organic molecules and sulfuric acid, in bright green, organic molecules alone, sulfuric acid, and ammonia, and sulfuric acid alone. The pie chart shows annual averages below 5 km altitude. Solid lines bound the ion-induced fraction, so in the present day, atmosphere around half of all NPF was ion-induced, and at this altitude, neutral nucleation of sulfuric acid alone and neutral pure organic nucleation are negligible. Adapted from Gordon et al. (2017).

activation and kinetic NPF schemes were used. The same model over a larger domain found that in China, NPF contributes 26% of CCN number at 1% supersaturation despite the very high emissions of primary particles (Matsui et al., 2013). In Europe, similar studies with PMCAMx (Fountoukis et al., 2012) and GLOMAP (Reddington et al., 2011) suggest NPF is more important, increasing concentrations of particles with at least 50 nm diameter by as much as a factor of 2. Julin et al. (2018) used PMCAMx with nucleation mechanisms involving both ammonia and amines to predict the consequences of future emission controls in Europe. A more focused study used the Consortium for Small-scale Modelling-Aerosols and Reactive Trace gases regional model COSMO-ART to show that NPF is enhanced in the plumes of "clean" power plants with reduced sulfur and fly ash emissions (Junkermann et al., 2011). The localized NPF in power plant plumes was also parameterized for large-scale models by Stevens and Pierce (2013) using large eddy simulations. The nucleation mechanisms in the PMCAMx study (Fountoukis et al., 2012) highlight the importance of ammonia in boundary layer nucleation, while those of the GLOMAP study highlight the importance of organics. A nucleation parameterization including the synergy between these compounds is a priority for future research.

5.8. Influence of NPF on CCN Concentrations and Climate

Globally, the indirect radiative effect of NPF has been studied in several models, and estimates vary widely even for individual nucleation processes. Kazil et al. (2010) found the radiative effect of ion nucleation was 1.15 W/m^2, while F. Yu, Luo, et al. (2012) calculate -3.67 W/m^2. The variation is not surprising, because the radiative effect of nucleation depends on uncertain vapor concentrations, uncertain aerosol-cloud interactions, and uncertain concentrations of the preexisting particles that form the *CS*, as well as the uncertainties in the nucleation parameterizations themselves. The radiative effect of BVOCs was not considered in these early studies but also depends strongly on whether or not they are allowed to participate in NPF in the boundary layer (Scott et al., 2014). The inclusion of biogenic NPF in models reduces estimates of indirect forcing (Gordon et al., 2016), while increased ammonia concentrations since preindustrial may also contribute a significant forcing via their role in NPF (Dunne et al., 2016).

Our understanding of NPF in the preindustrial atmosphere is especially uncertain. Without organics compounds participating in nucleation, Pierce and Adams (2009c) calculate an overall increase in CCN concentrations over the industrial period of 220%, due partly to primary emissions and partly to nucleation. With organic compounds nucleating with and without sulfuric acid, this increase was reduced to only 60% (Gordon et al., 2016), though some of the difference between these two results may be unrelated to nucleation as a different model was used. The uncertainty in preindustrial baseline CCN concentrations, for example, due to fire emissions (Hamilton et al., 2018) may mean these estimated increases change again. As the full ensemble of model uncertainties are better understood, we speculate that NPF will likely play an important role in future refinements to aerosol forcing estimates.

Compared to changes to precursor vapor emissions, changes in atmospheric ion concentrations in recent history have been very small relative to changes in gas concentrations from anthropogenic emissions or volcanic eruptions, of order 20% since the Maunder Minimum (Usoskin et al., 2015). Aerosol microphysics

models were used to show that variations in ion concentrations over the solar cycle or on centennial time scales (Svensmark & FriisChristensen, 1997) are very unlikely to substantially impact present day climate via nucleation because of this small variability (Dunne et al., 2012; Kazil et al., 2006; Kazil et al., 2012; Pierce & Adams, 2009a; Yu & Luo, 2014c). However, ions are still important. Based on CLOUD measurements, Gordon et al. (2017) calculate that 50% of particle formation involves ions. Before the industrial pollution, this figure was likely even higher, at an estimated 59%. Ions may also have a marginal effect on the *GR* of new particles (Laakso et al., 2003), but this process was considered in a sensitivity study by Snow-Kropla et al. (2011) and found to be negligible. However, the role of ions in paleoclimatic variability, or in the atmospheres of other planets, remains a promising avenue for further investigation.

The new generation of Earth system models are ideally suited to study feedbacks from temperature and land use changes involving NPF, especially cloud feedbacks and effects on NPF precursor emissions. Increasing biogenic emissions in a warmer climate may lead to increased nucleation and growth (Paasonen et al., 2013; Scott, Arnold, et al., 2018), while deforestation-related decreases in biogenic emissions may lead to reduced nucleation and hence climate warming (Scott, Monks, et al., 2018), but the large uncertainty may mean cooling is also possible (Hantson et al., 2017).

The Arctic region can be singled out as NPF in the Arctic is of particular interest for climate, as *CS* in the region are low and the clouds are very sensitive to aerosol (Mauritsen et al., 2011). While sea-ice loss was found not to lead to a large negative radiative feedback due to increased NPF from DMS, radiative effects in this region from NPF can be substantial (Browse et al., 2014). Using Goddard Earth Observing System-Chem GEOSchem TOMAS, Croft, Wentworth, et al. (2016) calculate a 0.5 W/m^2 cooling averaged over the Arctic due to the enhancement of NPF by ammonia from seabird guano.

5.9. Cloud Feedback on NPF

NPF near clouds, or in the outflow of deep convective clouds, is a well-established phenomenon (Clarke et al., 1998; Hegg et al., 1990; Lee et al., 2004; Weber et al., 1999). While some NPF can be captured in modeling studies using standard binary nucleation parameterizations (Clarke et al., 1999), clouds motivate new studies of NPF as the spatial resolution of regional climate models increases. Because NPF rates are nonlinear in precursor gas concentrations, which are spatially inhomogeneous near clouds, higher resolution reduces averaging biases. The NPF observed inside pockets of open cells generated by scavenging of aerosol was captured by a high-resolution model (Kazil et al., 2011). The role of clouds in the NPF process can now be studied with new aerosol-coupled large eddy simulations (Tonttila et al., 2017).

Clouds will strongly affect the balance of nucleation precursors by scavenging the most soluble gases. Therefore, water-insoluble organic vapors such as monoterpenes may be important for NPF close to clouds as well as SO$_2$ (Kulmala et al., 2006). The Stockholm cloud-resolving model is able to simulate inorganic (Ekman et al., 2008) and organic NPF inside convective clouds (Murphy et al., 2015). It was tested over the Amazon with the nucleation parameterization schemes of Metzger et al. (2010) and a homogeneous nucleation mechanism for pure organic nucleation based on CNT (Seinfeld & Pandis, 2016). Only the homogeneous organic nucleation could reproduce the observations of the Large-Scale Biosphere-Atmosphere – Cooperative Airborne Regional Experiment LBA-CLAIRE campaign (Krejci et al., 2003).

6. Summary: Outstanding Questions, Challenges, and New Directions

In the last decade, the greatest achievement in the field of NPF has been the beginnings of a quantitative understanding of the role of molecules other than sulfuric acid in the NPF process. This advance, which is still very much work in progress, has been made possible primarily by advances in instrumentation, especially the proliferation and improvement of mass spectrometers able to measure nucleated clusters, the development of appropriate ion source technologies for these instruments, and the development of more accurate counters for the smallest newly formed particles. The use of HR-TOF-MS has made it possible to identify some of the organic molecules which are likely to form particles, both in laboratory and field studies. Critically important, the operation of new facilities, especially the CLOUD laboratory, has enabled neutral and ion nucleation of specific precursor gas mixtures to be quantified separately in almost contaminant-free conditions.

In parallel, advances in quantum chemistry have facilitated significant progress in describing nucleation accurately. Both the experimental and theoretical advances are rapidly being translated to new

understanding of molecular clustering in ever more complicated systems and propagated to atmospheric models via parameterizations. Field campaigns and long-term measurements in more diverse environments have provided substantial additional insights, as well as a critically important and ever-growing database of observations for model evaluation. Meanwhile, explicit representations of NPF have been included in more diverse atmospheric and Earth system models, leading to new opportunities to study the important role of NPF in Earth's changing atmosphere, from the scale of individual clouds to that of the globe.

Despite these new findings, there still exists inadequate knowledge concerning the fundamental chemical mechanisms leading to nucleation and growth of nanoparticles. Available theories still cannot account for NPF events observed under diverse tropospheric conditions, and the identities of the chemical species contributing to nucleation and growth of nanoparticles have yet to be unambiguously established. Also, results of recent laboratory experiments, theoretical calculations, and field measurements are often controversial concerning the detailed roles of inorganic acids (i.e., sulfuric acid), base species (i.e., ammonia and amines), organic compounds (i.e., organic acids, oligomers, and HOMs), and ions as well as the synergy of these various species in promoting NPF as discussed in previous sections. Furthermore, the atmospheric conditions regulating NPF events are also poorly understood, including temperature, RH, and preexisting background aerosols.

One of the greatest challenges in laboratory experiments and field measurements of NPF lies in the lack of chemical speciation during nucleation and growth of nanoparticles. Although major analytical advances have been made in recent years to detect gaseous precursors and neutral and ionic prenucleation clusters, especially with the development of CIMS and CI-APi-TOF, there still exist major drawbacks for the available instruments to detect the NPF precursor species. For example, the current mass spectrometer-based techniques do not differentiate isomers (i.e., species with identical molecular weight but with different molecular structures). Also, even with the soft ionization in the application of CIMS, fragmentation still occurs. In addition, detection of neutral/ionic clusters is further complicated due to the effects of cluster growth (by adding monomers via ion-molecule reactions) or decomposition (by losing monomers) within the mass spectrometer system, yielding chemical compositions that differ considerably from those of ambient clusters. In addition, HOMs have been detected in numerous laboratory experiments/field measurements and in the gaseous/condensed phases using CI-APi-TOF, but they are still generically characterized only by volatility. True molecular identities of HOMs are yet to be elucidated to establish their chemical reactivity and to assess their roles in NPF. Furthermore, analytical methods have yet to be developed to measure the chemical composition of particles in the size range from 3 to 20 nm.

Currently, experimental results on the physiochemical properties of subnanometer-sized particles are lacking, even though these properties for nanoparticles are distinct from those of the bulk phases. Specifically, the density, surface tension, volatility, and hygroscopicity of nanoparticles have yet to be measured for atmospherically relevant species, including sulfuric acid, ammonia/amines, organic acids/oligomers/HOMs, and ions as well as their mixtures. Those quantities are necessary to validate atmospheric models that typically correct for the curvature effects employing an empirical formulation (i.e., the nano-Köhler theory). Also, recent analytical advance has extended the detection of particle number and size distribution of nanoparticles down to the nanometer range with the development of nano-DMAs, DEG-CPC, and PSM. However, the applications of particle counters are likely biased with respect to the volatility and composition of nanoparticles. Such artifacts can have direct consequences on both field studies and laboratory nucleation experiments, which typically characterize the nucleation efficiency of chemical species (i.e., organic compounds and ammonia/amines) relative to a reference system (i.e., the sulfuric acid-water binary nucleation) by an enhancement factor determined from the measured J.

Furthermore, available nucleation experimental studies using environmental chambers typically simulate the photooxidation of single compound or a mixture of a few compounds; extrapolation of the laboratory results to atmospheric conditions is highly uncertain. Even for the state-of-the-art CLOUD facility at CERN with the advantage of high purity, it is still challenging to replicate realistic ambient chemical complicity for NPF events under pristine and polluted environments. Multiple inorganic and organic species always coexist in the atmosphere, and the synergetic effects of photochemical oxidation of multiple inorganic and organic compounds likely enhance or suppress NPF.

Recently improved power in supercomputation has tremendously facilitated theoretical investigation for prenucleation clusters using quantum chemical methods and molecular dynamic simulations. However, the available theoretical methods are still unable to predict the nucleation barrier on the basis of the

potential energy surface during NPF and to estimate the size and chemical composition of the critical nucleus. For large multicomponent molecular clusters, obtaining reliable thermodynamic data is still the bottleneck, requiring advanced structure search algorithms (Lin et al., 2018; Xu & Zhang, 2012; Xu & Zhang, 2013). Theoretical investigation of aerosol nucleation relies mostly on the cluster stability. The thermodynamic data for stable clusters predicted from quantum chemical calculations are typically employed as the inputs for cluster dynamical models (e.g., ACDC) simulations to derive time-dependent evolution for molecular cluster concentrations. There exist additional deficiencies in the available cluster dynamical models, attributed to several inherent assumptions. For example, the forward reaction rate constants (i.e., k_i^+ in equation (5)) using the cluster dynamical model are commonly assumed to be equal to the collision coefficients and are calculated as hard-sphere collision rates based on the cluster radii. Also, the backward rate constants (evaporation coefficients or k_i^- in equation (5)) are computed using the Gibbs free energies of formation of clusters under an equilibrium condition. However, steady-state equilibrium for prenucleation clusters is rarely established under atmospheric conditions, because of continuous forward reactions by adding monomers to form larger clusters during NPF. In reality, the likelihood that a cluster grows to form a nanometer size particle or decomposes back to gaseous molecules is dependent on the competition between the forward reaction by adding a monomer and the backward reaction by losing a monomer (evaporation) at each intermediate step of the cluster growth. While the evaporation rate is dependent on the thermodynamic stability of the cluster, the forward rate constant is kinetically controlled, dependent on the activation and kinetic energies of the colliding clusters and monomers. Electrostatic attraction between ionic clusters and polar molecules and dipole-dipole interaction for neutral clusters clearly plays a key role in reducing the activation barrier.

Those above-mentioned deficiencies in laboratory experiments, theoretical calculations, and field measurements considerably hinder the effort in developing predictive parameterizations to account for the NPF process in atmospheric models. In particular, the diversity of organic compounds produced from photooxidation of biogenic and anthropogenic hydrocarbons and the diversity of amines and other species that may contribute to NPF make it very difficult to assess their contributions to nucleation and growth of nanoparticles. A possible way forward is to represent and parameterize the detailed chemistry in box modeling studies, using simplifying approaches such as VBS. Box model studies can be used to derive the simplifications are needed for climate models, where computing limitations dictate that only a few precursor vapors for NPF can be represented explicitly. However, the difficulties involved in accurately compiling emission inventories for NPF precursors are likely to place a hard limit on the accuracy with which NPF can be modeled, irrespective of the accuracy with which the physical and chemical processes involved in NPF itself are represented.

As a critical source of the tropospheric aerosol population, NPF needs to be understood at the fundamental molecular level. Further laboratory experiments and field measurements are necessary to improve chemical speciation for nucleation and growth of nanoparticles, including simultaneous detection of gaseous nucleating vapors and chemical compositions for neutral/ionic clusters and nanoparticles. To achieve this level of chemical speciation, advanced analytical instruments need to be developed to identify and quantify the gaseous nucleating precursors and to measure the chemical composition from prenucleation clusters, critical nucleus, to nanoparticles. Novel experimental methods are also necessary to investigate NPF under conditions that mimic ambient chemical complexity (i.e., atmospherically relevant types and abundances for chemical species), temperature, RH, preexisting particles, and time scales. Advanced theoretical methodologies need to be developed to interpret and corroborate the findings of laboratory experiments and field measurements. These approaches are necessary to reliably predict thermodynamic data for large multicomponent molecular clusters, accurately account for the detailed kinetics for the forward and backward reactions at each stage of the cluster growth, and efficiently describe the activation barrier and the size and chemical composition of the critical nucleus. Furthermore, physically based parametrizations of aerosol nucleation and growth need to be established on the basis of improved laboratory, field, and theoretical studies and to be incorporated into atmospheric models to assess the impacts of aerosols on human health, weather, and climate.

Appendix A: List of Acronyms

ACDC   Atmospheric Cluster Dynamics Code
AIS   Air ion spectrometer

APi-TOF	Atmospheric pressure ionization-time of flight mass spectrometer
APM	Advance particle microphysics
AVOC	Anthropogenic volatile organic compound
BVOC	Biogenic volatile organic compound
CCN	Cloud condensation nuclei
CERN	European Organization for Nuclear Research Conseil européen pour la recherche nucléaire
CI-APi-TOF	Chemical ionization atmospheric pressure ionization-time of flight mass spectrometer
CIMS	Chemical ionization mass spectrometer
CLOUD	Cosmics Leaving Outdoor Droplets
CNT	Classical nucleation theory
COSMO-ART	Consortium for Small-Scale Modeling-Aerosols and Reactive Trace Gases
CARMA	Community Aerosol and Radiation Model for Atmospheres
CPC	Condensation particle counter
CS	Condensation sink
DEG	Diethylene glycol
DMA	Dimethylamine Differential mobility analyzer
DMS	Dimethyl sulfide
ELVOC	Extremely low volatile organic compound
GEOS-chem	Goddard Earth Observing System-Chem
GLOMAP	Global Model of Aerosol Processes
GR	Growth rate
HOM	Highly oxidized molecule
HOM-ONs	Organonitrate highly oxidized molecules
HR-DMA	High-resolution DMA
HR-TOF-MS	High-resolution time of flight mass spectrometry
IIN	Ion-induced nucleation
IMN	Ion-mediated nucleation
LBA-CLAIRE	Large-Scale Biosphere-atmosphere-Cooperative LBA Airborne Regional Experiment
LVOC	Low volatile organic compound
MSA	Methanesulfonic acid
NAAMES	North Atlantic Aerosols and Marine Ecosystems Study
NAIS	Neutral Cluster and Air Ion Spectrometer
NPF	New particle formation
PM	Particulate matter
PMCAMx	Particulate Matter Comprehensive Air Quality Model
PMCAMx-UF	Particulate Matter Comprehensive Air Quality Model with Extensions-Ultrafine
PSM	Particle size magnifier
RH	Relative humidity
SAGE II	Stratospheric Aerosol and Gas Experiment
SAWNUC	Sulfuric-Acid-Water Nucleation
SMEAR II	Station for Measuring Forest Ecosystem-Atmosphere Relations
SMPS	Scanning mobility particle sizer
SVOC	Semi-volatile organic compound
TOMAS	TwO-Moment Aerosol Sectional
VBS	Volatility Basis Set
VOC	Volatile organic compound
WRF	Weather Research Forecasting
WRF-MOSAIC	Weather Research and Forecasting-Model for Simulating Aerosol Interactions and Chemistry

Table B1

Parameters in the NPF Model

Parameter	Value	Parameter	Value
pb, n	3.95451	pb, i	3.373738
ub, n	9.702973	ub, i	-11.48166
vb, n	12.62259	vb, i	25.49469
wb, n	-0.007066146	wb, i	0.1810722
pt, n	2.891024	pt, i	3.138719
ut, n	182.4495	ut, i	-23.8002
vt, n	1.203451	vt, i	37.03029
wt, n	-4.188065	wt, i	0.227413
pA, n	8.003471	pA, i	3.071246
a_n	1.5703478×10^{-6}	a_i	0.00483140
k_{SA-Org}	3.27×10^{-21}	a_1	0.0400097
a_2	1.84826	a_3	0.00136641
a_4	1.56588	a_5	0.186303

Note. The full numerical precision is needed. The units of [HOM] are 10^7 cm^{-3}, [H$_2$SO$_4$] has units 10^6 cm^{-3} in the inorganic parametrization (J_{SA} and $J_{SA,NH3}$) and cm^{-3} in J_{SA-org}, and [NH$_3$] has units 10^6 cm^{-3}. The ion and BioOxOrg concentrations have unit cm^{-3}.

Appendix B: Nucleation and Growth Parameterizations Derived From CLOUD Experiments

The full NPF model from the CLOUD experiment (Gordon et al., 2017) consists of the sum of J_{SA}, J_{SA-NH3}, J_{SA-org}, and J_{org}. The inorganic components are given by (Dunne et al., 2016)

$$J_{SA} = k_{b,n}(T)[\text{H}_2\text{SO}_4]^{P_{b,n}} + k_{b,i}(T)[\text{H}_2\text{SO}_4]^{P_{b,i}}[n_-],$$

and

$$J_{SA,NH3} = k_{t,n}(T)f_n([\text{NH}_3], [\text{H}_2\text{SO}_4])[\text{H}_2\text{SO}_4]^{p_{t,n}}$$
$$+ k_{t,i}(T)f_i([\text{NH}_3], [\text{H}_2\text{SO}_4])[\text{H}_2\text{SO}_4]^{p_{t,i}}[n_-],$$

where all the $k(T)$, for temperatures T in Kelvin, are given by

$$\ln k_{(x,y)} = u_{(x,y)} - \exp\left(v_{(x,y)}\left(\frac{T}{1,000} - w_{(x,y)}\right)\right)$$

where $x = b$ or t (binary and ternary) and $y = n$ or i (neutral and ion-induced). The ion concentration is $[n_-]$. The functions $f_y([\text{NH}_3], [\text{H}_2\text{SO}_4])$ are

$$f_y([\text{NH}_3], [\text{H}_2\text{SO}_4]) = \frac{[\text{NH}_3]}{a_y + \frac{[\text{H}_2\text{SO}_4]^{p_{t,y}}}{[\text{NH}_3]^{p_{A,y}}}}.$$

The organic components are (Kirkby et al., 2016; Riccobono et al., 2014)

$$J_{SA-org} = 0.5 k_{SA-org}[\text{H}_2\text{SO}_4]^2[\text{BioOxOrg}],$$

$$J_{org} = J_n + J_{iin},$$

$$J_n = a_1[\text{HOM}]^{a_2 + a_5/[\text{HOM}]},$$

$$J_{iin} = a_3[\text{HOM}]^{a_4 + a_5/[\text{HOM}]}[n_\pm].$$

The values of the parameters are given in Table B1; attention must be paid to the units of the gas concentrations, specified in the caption.

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