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TOPICAL REVIEW

Atmospheric new particle formation and growth: review of field observations

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Abstract

This review focuses on the observed characteristics of atmospheric new particle formation (NPF) in different environments of the global troposphere. After a short introduction, we will present a theoretical background that discusses the methods used to analyze measurement data on atmospheric NPF and the associated terminology. We will update on our current understanding of regional NPF, i.e. NPF taking simultaneously place over large spatial scales, and complement that with a full review on reported NPF and growth rates during regional NPF events. We will shortly review atmospheric NPF taking place at sub-regional scales. Since the growth of newly-formed particles into larger sizes is of great current interest, we will briefly discuss our observation-based understanding on which gaseous compounds contribute to the growth of newly-formed particles, and what implications this will have on atmospheric cloud condensation nuclei formation. We will finish the review with a summary of our main findings and future outlook that outlines the remaining research questions and needs for additional measurements.

1. Introduction

Atmospheric new particle formation (NPF) and growth involves the formation of molecular clusters and their subsequent growth to larger sizes, first to a few nm in particle diameter, then to nucleation and Aitken mode particles in the sub-100 nm size range, and possibly up to sizes at which these particles may act as cloud condensation nuclei (CCN). According to our current understanding, molecular cluster formation appears to take place almost everywhere and all the time in the atmosphere, whereas the formation of growing nanoparticles either by homogeneous or heterogeneous nucleation requires more specific atmospheric conditions (Kulmala *et al* 2014). Simulations using different large-scale modeling frameworks and different parameterizations for this phenomenon suggest that NPF is the dominant source of the total particle number concentration, and an important contributor to the CCN budget, in the global troposphere (Spracklen *et al* 2006, Merikanto *et al* 2009, Pierce and Adams 2009, Yu and Luo 2009, Makkonen *et al* 2012, Dunne *et al* 2016, Gordon *et al* 2017) as well as in the continental boundary layer (BL) (Reddington *et al* 2011, Fountoukis *et al* 2012, Matsui *et al* 2013, Lupascu *et al* 2015, Posner and Pandis 2015, Cai *et al* 2016). The situation in different atmospheric environments is more diverse, and poorly quantified, at the moment. This feature seriously hinders our ability to estimate the climate and health effects of atmospheric aerosol particles, and especially the role of human actions in those effects.

During the past decade or so, a number of scientific reviews, or compilation studies, on atmospheric NPF has been written (Kulmala *et al* 2004, O'Dowd and Hoffmann 2005, Curtius 2006, Holmes 2007, Enghoff and Svensmark 2008, Kazil *et al* 2008, Kulmala and Kerminen 2008, Hegg and Baker 2009, Bzdek and Johnston 2010, Kerminen *et al* 2010, Hirsikko *et al* 2011, Kulmala *et al* 2012, Vehkamäki and Riipinen 2012, Zhang *et al* 2012, Kulmala *et al* 2014, Li *et al* 2015b, Kulmala *et al* 2016b, Wang *et al* 2017a, Nieminen *et al* 2018). These papers have focused on varying aspects of atmospheric NPF, typically covering one or more of the following topics: (1) the observed character of NPF in different atmospheric environments, including the particle formation and growth rates (GRs) during NPF and frequency at which NPF occurs, (2) the chemistry of atmospheric NPF, (3) the thermodynamics and kinetics of NPF, (4) atmospheric NPF mechanisms, including the role of ions in this process, (5) analysis of the factors favoring, or disfavoring, atmospheric NPF, and (6) instrumental issues related to investigating NPF.

In the review presented here, we will focus on the observed characteristics of atmospheric NPF in different tropospheric environments. There are several reasons for doing that. First, no comprehensive review on this topic has been published since Kulmala *et al* (2004). Second, there has been plenty of work in this research field during the past few years, with a large number of new observational results published in different scientific journals. Third, the published work on atmospheric NPF relies on a vast variety of different approaches that have not been properly reviewed, or even discussed, earlier. Fourth, while it has become obvious that atmospheric NPF occurs frequently in very different tropospheric environments ranging from remote polar areas to heavily-polluted megacities, only few comparisons between contrasting environments in terms of atmospheric NPF have been conducted. Finally, the scientists dealing with atmospheric NPF, including those working with large-scale atmospheric models and those making laboratory experiments or field measurements, are clearly in need for the kind of information to be discussed in this review.

Since the methodology used to analyze measurement data on atmospheric NPF, and even the associated terminology, varies greatly between the individual studies, we will first present a theoretical background that discusses these issues (section 2). After that, we will provide a short update on our current understanding of regional NPF, i.e. NPF taking place simultaneously place over spatial scales of tens to hundreds of km, and complement that with a full review on reported NPF and GRs during regional NPF, along with reported NPF event frequencies (section 3). Atmospheric NPF taking place at sub-regional scales will be reviewed in section 4. Since the growth of newly-formed particles into larger sizes is of great current interest, we will also discuss our observation-based understanding on which gaseous compound contribute to the growth of newly-formed particles, and what implications this will have on atmospheric CCN formation (section 5). We will end this paper with a future outlook that discusses the remaining research questions and needs for additional measurements (section 6).

The nucleation mechanism and initial steps of atmospheric NPF is left out from this paper because this topic was reviewed relatively recently (Kulmala *et al* 2014), and because this research branch is under a very fast development phase at the moment. Partly for the same reason, we constrain our analysis to sizes larger than about 3 nm in particle diameter, unless otherwise mentioned. This constraint is important to the theoretical background presented in section 2, since many of the methods and concepts discussed in that section become increasingly inaccurate at particle diameters smaller than 2–3 nm. In order to keep the length of this paper within reasonable limits, we will not discuss the rich instrumentation currently applied in studying atmospheric NPF. Modeling and laboratory studies on NPF will be mentioned only if needed for complementing the analysis presented below.

2. Theoretical background

2.1. Identifying, characterizing and classifying atmospheric NPF

From the observational point of view, atmospheric NPF and subsequent particle growth are seen as an emergence of new aerosol particles into the lower end of the measured particle size spectrum, followed by the growth of these particles into larger sizes. If this phenomenon is taking place regionally, i.e. over a minimum distance of a few tens of km in radius, a contour plot displaying measured particle number size distributions as a function of time at a fixed location often reminds a banana (figure 1(a)). These so-called ‘banana plots’ are idealizations of real NPF and growth taking place in the atmosphere: any inhomogeneity in the measured air masses, and more specifically in processes modifying particle number size distributions in these air masses, causes irregularities in the shapes of banana-like features. When NPF and particle growth are taking place sub-regionally, i.e. over distances of a few km or less, measurements at a fixed location tend to capture only a limited part of this process. In a contour plot displaying measured particle number size distributions as a function of time, NPF taking place in sub-regional scales may be visualized in a variety of shapes, one example of which is illustrated in figure 1(b).

When estimating the importance of atmospheric NPF in different environments, one needs to know how frequently this phenomenon occurs and how strong it is when taking place. A useful concept in this regard is the co-called ‘NPF event’ which starts when NPF is first observed to take place and ends when no more new particles enter the measured particle size range (see figure 1(a)). The number of individual NPF

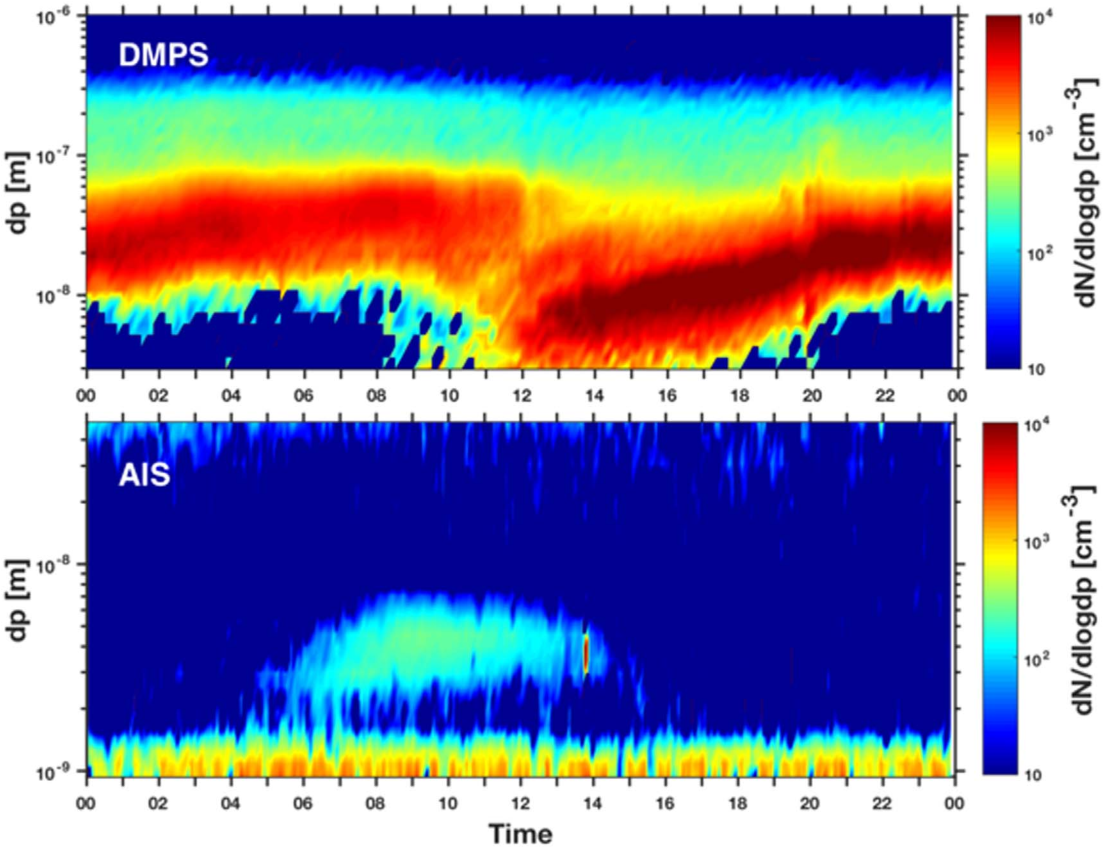

Figure 1. (Top) An example of a regional NPF event day, as measured by a differential particle mobility sizer (DMPS) in Hyytiälä, Finland, on 8 April 2017. The data is presented in UTC + 2 (Finnish winter time). The time in the *x*-axis refers to the time of day, whereas the color indicates the number density of particles of different size (particle diameter, *y*-axis). One may see that the newly-formed particles appear into the measured size distribution at around the noon, after which they grow in size reaching diameters between about 15 and 50 nm by midnight. (Bottom) An example of a local NPF event day, as measured by an air ion spectrometer (AIS) at Dome C, Antarctica, on 19 March 2011. The data are presented in UTC (local time in UTC + 8). Unlike the feature in the regional NPF event, no clear growth in size is observed in the local NPF event after the appearance of a new mode between 2 and 10 nm in the measured size distribution. The presence of this new mode lasts for several hours.

events recorded over a longer period of time (e.g. month, year) then defines the frequency of atmospheric NPF. In case of regional NPF, one rarely observes more than one NPF event per day (see section 3.1.2 for a more detailed discussion), so a convenient way to express the frequency of NPF is to calculate the fraction of days having a NPF event. The frequency of NPF is not a well-defined quantity for sub-regional NPF, since measurements conducted at a fixed location tend to capture a rather random subset of small-scale NPF events occurring upwind of the measurement site.

The strength of an individual NPF event is characterized by its duration, the rate at which new particles are formed during the event, and the GR of these particles into larger sizes. For regional NPF taking place homogeneously over large distances, all these quantities can be determined in a relatively straightforward manner from continuous particle number size distribution measurements (see section 2.3). In the real atmosphere, however, spatial heterogeneities in the processes affecting particle number size distribution may preclude a reliable determination of one or more of these quantities. Determining the strength of a NPF event becomes increasingly difficult when the spatial extent of this phenomenon gets smaller, so quantities like the particle formation and GR can only seldom be determined for sub-regional NPF events.

A proper analysis of NPF from atmospheric observations, as well as comparisons between different data sets and studies, require consistent criteria for identifying and classifying this phenomenon. In this regard, Dal Maso *et al* (2005) defined 'NPF event days' as those days during which (1) a distinct new mode of particles appears in the particle number size distribution, (2) this mode is located initially below 25 nm of the particle diameter, (3) the mode prevails for more than an hour, and (4) the mode shows signs of growth. The rest of the days were defined either as 'non-event days' during which no NPF was observed to take place, or 'undefined days' for which determining whether NPF had been taken place or not was ambiguous. According to Dal Maso *et al* (2005), NPF event days can be further categorized into sub-classes based on the amount and accuracy of information that could be derived to characterize the NPF event. Also undefined days can be divided into a few sub-classes based on the method introduced by Buenrostro Mazon *et al* (2009).

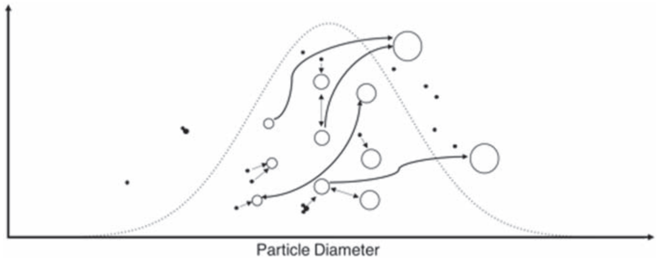

Figure 2. Growth of individual aerosol particles (open circles) and a particle population (all open circles inside the dashed region). Individual particles grow by the net flux of vapor molecules (black small circles) or their clusters into them, illustrated by thin one-headed arrows. Thick one-headed arrows represent the coagulation scavenging of particles in this population onto larger particles. The two-headed arrows represent self-coagulation within this particle population.

While the classification scheme introduced by Dal Maso *et al* (2005) and its later refinements (e.g. Kulmala *et al* 2012) have been widely used in analyzing various characteristics of regional-scale NPF, a proper application of this scheme requires continuous measurements of particle number size distributions over a relatively wide particle diameter range, preferably at a fixed location. Since such measurements are not always available, a number of alternative methods for identification or classification of atmospheric NPF events have been developed and applied in the scientific literature. These include approaches aiming to identify whether NPF had been occurred or not using total particle number concentration measurements in some specific size range using a moving platform (e.g. Stratmann *et al* 2003, Siebert *et al* 2004, Junkermann *et al* 2011a), and methods classifying NPF and growth events based on ion spectrometer measurements (e.g. Hirsikko *et al* 2007, Venzac *et al* 2007, Manninen *et al* 2010, Rose *et al* 2013, Leino *et al* 2016) or combinations of ion spectrometer and total particle measurements (e.g. Yli-Juuti *et al* 2009). In addition to these examples, different identification and classification criteria are commonly needed when analyzing NPF events that are spatially limited, or of short duration, and in cases when the growth of newly-formed particles to larger sizes is substantially suppressed.

2.2. Quantities relevant for analyzing NPF and GRs

The two most important quantities describing atmospheric NPF events are the particle formation rate (J) and the particle GR. In the scientific literature, somewhat different definitions for both J and GR can be found depending on (1) whether a microscopic or macroscopic approach to analyzing NPF and growth is assumed, and (2) which kind of experimental information is available for determining these two quantities. The microscopic approach considers explicitly the dynamics of molecular clusters of different sizes using a kinetic equation to describe the collision of molecules (monomers) with the clusters and the evaporation of monomers from them, whereas the macroscopic approach is based on the assumption that the particle number size distribution can be approximated to be a continuous function of the particle size (Holten and van Dongen 2009). In this paper, we will adopt the macroscopic approach to characterize the NPF and growth process. While this approach becomes highly inaccurate when approaching the sizes at which particles or clusters contain only few molecules (e.g. Wang *et al* 2013, Olenius *et al* 2015), it appears to perform reasonably well for the >3 nm particle diameter range focused on here.

2.2.1. Definition of relevant quantities

In the macroscopic view of NPF, the particle formation rate is defined as the flux of growing nanoparticles through a certain particle size barrier, described usually by the particle diameter, dp. This flux is denoted as J_{dp} , and a widely-used unit for it is $\mathrm{cm}^{-3}\,\mathrm{s}^{-1}$ (or particles $\mathrm{cm}^{-3}\,\mathrm{s}^{-1}$). Two other quantities related to the particle formation rate are the clustering rate and nucleation rate. The clustering rate refers to the net formation rate of small clusters consisting of a small number of cluster building blocks, usually molecules, being an essential quantity in the microscopic description of NPF. In case there is a well-defined energy barrier that the clusters need to overcome in order to produce growing nanoparticles, the formation rate of clusters passing this barrier is called the nucleation rate. In the scientific literature, the terms particle formation rate and nucleation rate are used sometimes synonymously, even though it should be kept in mind they are two separate theoretical concepts that should not be mixed with each other.

When analyzing particle GRs, it is important to distinguish between the growth of an individual aerosol particle and the growth of a particle population (figure 2). The conservation of mass defines directly the GR of an individual aerosol particle, $\mathrm{GR}_{\mathrm{ind}} = d\mathrm{dp}/dt$, as:

$$\frac{dm_{\mathrm{p}}}{dt} = \frac{d(\rho_{\mathrm{p}}V_{\mathrm{p}})}{dt} = V_{\mathrm{p}}\frac{d\rho_{\mathrm{p}}}{dt} + \frac{1}{2}\pi\,\rho_p\mathrm{dp}^2 \times \frac{d\mathrm{dp}}{dt} \approx \frac{1}{2}\pi\,\rho_{\mathrm{p}}\mathrm{dp}^2\mathrm{GR}_{\mathrm{ind}}. \qquad (1)$$

Here m_{p}, V_{p} and ρ_{p} are the particle mass, volume and density, respectively, t is the time, and the term

dm_p/dt represents the net mass flux of new material into the particle. The last form of the equation includes the approximation that the addition of new mass into the particle changes little the particle density. The net mass flux into the particle can be caused by condensation of low-volatile (LVOC) vapors into the particle, by heterogeneous formation of low-volatility substances on the surface or inside the particle from more volatile vapors, or by coagulation of small clusters with the particle (Seinfeld and Pandis 1998, Riipinen *et al* 2012, Lehtipalo *et al* 2016).

By a growing particle population we mean a reasonably well-separated mode of particles undergoing a growth into larger sizes, typically nucleation or Aitken mode particles formed during the past few hours by NPF. The mean size of such a mode increases not only by the growth of individual aerosol particles ($\mathrm{GR_{ind}}$), but also by self-coagulation ($\mathrm{GR_{scoag}}$) and coagulation scavenging ($\mathrm{GR_{scav}}$). In self-coagulation, some of the particles inside the growing mode collide with each other (figure 2), which increases the mean size of the mode and simultaneously decreases the total particle number concentration of this mode. In coagulation scavenging, some of the particles in the growing mode collide with larger particles, i.e. with particles clearly outside the growing mode (figure 2). Since such collisions are most efficient for the smallest particles in the growing mode (see section 2.2.2), this process increases the mean size of the mode although no real particle growth is taking place inside the mode. The overall GR of the mode is roughly the sum of the three contributions mentioned above

$$\mathrm{GR} \approx \mathrm{GR_{ind}} + \mathrm{GR_{scoag}} + \mathrm{GR_{scav}}. \tag{2}$$

Determining the value of each term in in equation (2), and especially separating the contribution of $\mathrm{GR_{scoag}}$ from those of $\mathrm{GR_{ind}}$ and $\mathrm{GR_{scav}}$, requires knowledge about the lower and upper borders of the growing particle mode. Determining this information can be very difficult for atmospheric particle number size distributions. The contribution of $\mathrm{GR_{scoag}}$ to GR is only important at very high concentrations ($>10^5\ \mathrm{cm^{-3}}$) of particles in the growing mode (Kerminen *et al* 2004, Anttila *et al* 2010). The most practical unit of GR in atmospheric applications is $\mathrm{nm\ h^{-1}}$.

Two additional concepts that are commonly used in analyzing atmospheric NPF are condensation sink (CS) and coagulation sink (CoagS) defined by Kulmala *et al* (2001):

$$\mathrm{CS} = 2\pi D \int_0^\infty \beta(\mathrm{dp}) \times \mathrm{dp}\; n(\mathrm{dp})d\mathrm{dp}, \tag{3}$$

$$\mathrm{CoagS(dp)} = \int_0^\infty K(\mathrm{dp}, \mathrm{dp}')n(\mathrm{dp}')d\mathrm{dp}'. \tag{4}$$

Here β(dp) is the transition-regime correction factor for condensation onto a particle with a diameter of dp (Fuchs and Sutugin 1971), D is the vapor diffusion coefficient, n(dp) is the continuous particle number size distribution function, and K(dp, dp′) is the Brownian coagulation coefficient between particles of diameters dp and dp′. The CS represents the average rate at which a non-volatile gaseous compound condenses into the entirely aerosol particle population, so the inverse of CS is equal to the average lifetime of a non-volatile compound in the gas phase. Similarly, CoagS (dp) can be viewed as the average rate at which particles of a diameter dp coagulate with the whole particle population, and its inverse represents the average particle lifetime against coagulation scavenging. It should be noted that, due to limitations in atmospheric particle number size distribution measurements, the integration over particle diameters in equations (3) and (4) is always cut from both lower and upper ends of the particle size spectrum. Leaving out the particles smaller than a few nm in diameter is usually well justified because such particles rarely make important contributions to either CS or CoagS. Contrary to this, in environments influenced heavily by dust, sea salt or primary biogenic particles, particles larger than a few hundred nm in diameter could substantially increase both CS and CoagS.

When investigating the role of gas-phase chemistry in the particle formation and growth, useful concepts are the total concentration of non-volatile gaseous compounds, C, and their production rate in the gas phase, Q (e.g. Kulmala *et al* 2001). These two quantities are highly idealized descriptions of a real atmosphere, where a large number of compounds of different volatility is simultaneously present. In order to deal with this issue, Donahue *et al* (2011) suggested to group organic compounds into a few volatility classes to describe their gas-particle partitioning: extremely low-volatile (ELVOC), LVOC, semi-volatile, intermediate-volatile and volatile (VOC) organic compounds. In many applications, C can be approximated as the sum of gaseous sulfuric acid and ELVOC concentration, even though it should be kept in mind that none of the gaseous compounds present in the atmosphere is strictly non-volatile. Likewise, it has been shown that the growth of atmospheric aerosol particles is affected not only by the least volatile compounds, but also by low- and semi-volatile compounds (e.g. Tröstl *et al* 2016).

2.2.2. Connections between the quantities

The formation of new atmospheric aerosol particles and their growth to larger sizes are coupled closely to each other, as well as to the concentrations of aerosol precursor compounds and properties of a pre-existing particle population. Understanding these couplings is essential when analyzing NPF using atmospheric observations. In the following we will summarize some of the most important couplings between the relevant variables (J, GR, CS, CoagS, C, Q, particle number size distribution function), keeping in mind that the derived equations become increasingly inaccurate when approaching the cluster regime as discussed in the previous sub-section.

Mathematically, the particle formation and GRs are connected to each other via the continuous particle number size distribution function, $n(\mathrm{dp})$:

$$J_{\mathrm{dp}} = n(\mathrm{dp}) \times \mathrm{GR}(\mathrm{dp}). \tag{5}$$

The GR defined by equation (5) was recently named as the flux-equivalent GR, since the GR of a particle population can be determined in different ways (Kontkanen *et al* 2016b; see also section 2.3.2). It should be noted that equation (5) is strictly valid only when the particle number concentration around the size dp is conserved, i.e. when $n(\mathrm{dp})$ is affected solely by the growth of individual aerosol particles. In its present form, equation (5) should not be applied for cases in which either self-coagulation or coagulation scavenging give a large contribution to the growth of a particle population.

Condensation and coagulation sinks describe essentially the same phenomenon, the only difference being that in CS one of the colliding parties is a gas molecule while in CoagS it is a molecular cluster or small aerosol particle, the other colliding party being the pre-existing particle population in both cases. Lehtinen *et al* (2007) showed that the CoagSs of particles with diameters dp_1 and dp_2 can be related to each other via the following approximate formulae:

$$\mathrm{CoagS}(\mathrm{dp}_2) \approx \mathrm{CoagS}(\mathrm{dp}_1) \times \left(\frac{\mathrm{dp}_1}{\mathrm{dp}_2}\right)^m, \tag{6}$$

where the exponent m (typically in the range 1.6−1.8) depends on the shape of the pre-existing particle population. A direct consequence of equation (6), together with the similarity between the condensation loss of a gas molecule and coagulation scavenging of a small aerosol particle, is that also CS and CoagS are connected with each other:

$$\mathrm{CoagS}(\mathrm{dp}) \approx \mathrm{CS} \times \left(\frac{\mathrm{dg}}{\mathrm{dp}}\right)^m, \tag{7}$$

where dg is the diameter of the condensing gas molecule. In practical applications, CS is often calculated by assuming the condensing vapor to be sulfuric acid, in which case dg should be the diameter of a sulfuric acid hydrate ($\mathrm{dg} \approx 0.71$ nm, see Lehtinen *et al* 2007).

Equation (6) indicates that the probability by which an aerosol particle is lost by coagulation decreases relatively rapidly with an increasing particle size, so the survival probability of a growing particle is determined by the competition between its GR and its scavenging rate by coagulation with larger particles. For a growing particle population, the formation rates of different-size particles can be related to each other via (Lehtinen *et al* 2007):

$$J_{\mathrm{dp}_2} = J_{\mathrm{dp}_1} \exp\left(\frac{\mathrm{dp}_1}{m+1}\left[1 - \left(\frac{\mathrm{dp}_2}{\mathrm{dp}_1}\right)^{m+1}\right] \times \frac{\mathrm{CoagS}(\mathrm{dp}_1)}{\mathrm{GR}}\right), \tag{8}$$

where the competition between the particle growth and coagulation scavenging appears as the ratio CoagS/GR. Equation (8) assumes implicitly that the particle GR is constant over the diameter range $[\mathrm{dp}_1, \mathrm{dp}_2]$, and that GR is not affected by self-coagulation. The first of these assumptions is usually not valid (see section 5.1). Korhonen *et al* (2014) derived a modified version of equation (8), in which the particle GR is allowed to change linearly with an increasing particle size. Self-coagulation does not affect the accuracy of equation (8), except at high formation rates of new particles, and an iterative procedure that takes this into account the effect of self-coagulation was introduced by Anttila *et al* (2010). Based on equation (7), Kerminen and Kulmala (2002) showed that the relation between $J_{\mathrm{dp}1}$ and $J_{\mathrm{dp}2}$ can also be written using CS instead of CoagS. Finally, a generalized numerical method for relating $J_{\mathrm{dp}1}$ and $J_{\mathrm{dp}2}$ from experimental data was recently introduced by Kürten *et al* (2015).

The concentration of any non-volatile compound in the gas phase (C) is determined by the balance between its production rate and its loss rate by condensation into pre-existing particles:

$$\frac{dC}{dt} = Q - \mathrm{CS} \times C. \tag{9}$$

In the atmosphere, C evolves towards a pseudo-steady state given by:

$$C = \frac{Q}{\mathrm{CS}}. \tag{10}$$

The accuracy of equation (10) depends on the magnitude of CS and the rate at which Q is changing over time (Kerminen *et al* 2004). Excluding very clean environments (small CS) and environments with exceptionally rapid changes in Q (e.g. fresh pollution plumes), equation (10) can be considered as a good approximation to the value of C. Equation (10) offers a simple means to estimate C when this quantity cannot be measured directly. The application of this concept had led to the derivation of the widely-used proxy for the gas-phase sulfuric acid concentration (Petäjä *et al* 2009), and the recent proxy for the concentration of oxidized organic compounds in the gas phase (Kontkanen *et al* 2016c).

The GR of individual aerosol particles can be written as

$$\mathrm{GR}_{\mathrm{ind}} = \sum \mathrm{GR}_{\mathrm{cond},i} + \sum \mathrm{GR}_{\mathrm{het},i} + \mathrm{GR}_{\mathrm{oth}}, \tag{11}$$

where $\mathrm{GR}_{\mathrm{cond,i}}$ is the GR due to net condensation of vapor i into the particle, $\mathrm{GR}_{\mathrm{het},i}$ is the GR due to some heterogeneous reaction of vapor i on the surface or inside the particle, and $\mathrm{GR}_{\mathrm{oth}}$ represents other growth processes such as collisions of molecular clusters with the particle. The condensational growth term can further be written into the following form:

$$\mathrm{GR}_{\mathrm{cond},i} = A \times [C_i - C_{i,0} \times \mathrm{Ke(dp)}], \tag{12}$$

where C_i and $C_{i,0}$ are concentration of vapor i in the gas phase over a flat surface having the same composition as the particle, A takes into account the collision rate between vapor molecules and the particle, and Ke is the Kelvin term which increases rapidly with the decreasing particle size for particles smaller than a few nm in diameter (e.g. Wang *et al* 2013). For a non-volatile compound, $C_{i,0} = 0$ and A is relatively constant over the particle diameter range of 3–10 nm (Nieminen *et al* 2010), such that $\mathrm{GR}_{\mathrm{cond},i}$ is directly proportional to both C_i and Q_i (see equations (10) and (12)).This means that the contribution of any non-volatile compound to the particle GR can be estimated relatively easily once either its gas-phase concentration or its production rate in the gas-phase is known. In practical applications, sulfuric acid and ELVOCs can usually be treated as if they were non-volatile, whereas for more volatile vapors the term $C_{i,0} \times \mathrm{Ke(dp)}$ needs to be taken into account when estimating the value of $\mathrm{GR}_{\mathrm{cond},i}$.

2.3. Determining particle formation and GRs from atmospheric measurements

In the scientific literature, a large variety of methods have been developed and applied for determining the particle formation and GRs from measurement data. We will not make a full review on these methods here, but rather concentrate on cases where information on the time evolution of the particle number size distribution is available. The vast majority of the particle formation and GRs published so far are based on those kinds of measurements.

2.3.1. Particle formation rate

Let us consider a measurement device that provides data on particle number concentrations, N_i, in several size bins i over the diameter range $[\mathrm{dp}_{\min}, \mathrm{dp}_{\max}]$ and with a temporal resolution of Δt. From such measurements, one can determine the particle formation rate at the lower boundary of any of the size bin i, dp_i, that belongs to the diameter range $[\mathrm{dp}_{\min}, \mathrm{dp}_{\max}]$. This is in practice done by looking at the temporal behavior of the total particle number concentration over some number (n) of size bins starting from the bin i: $N = N_i + N_{i+1} + \ldots N_{i+n-1}$, where $\mathrm{dp}_{i+n} \leqslant \mathrm{dp}_{\max}$ (figure 3). The time evolution of N can be related to the particle formation rate at dp_i via the following balance equation:

$$\frac{\Delta N}{\Delta t} \approx J_{\mathrm{dp}_i} - J_{\mathrm{dp}_{i+n}} - \frac{1}{2}\sum_{j=i}^{i+n-1} K(\mathrm{dp}_j, \mathrm{dp}_j) N_j^2 - \sum_{j=i}^{i+n-1} \mathrm{CoagS}(\mathrm{dp}_j)_{>\mathrm{dp}_{j+1}} N_j + \sum_{j=i}^{i+n-1} P_j - \sum_{j=i}^{i+n-1} v_j N_j. \tag{13}$$

Here P_j is the rate of primary particle emissions into the size bin j, v_j is the loss rate of particles in this size bin by deposition, and the subscript $j + 1$ in CoagS means that only particles in the size bins $j + 1$, $j + 2 + \ldots$ are considered when calculating the CoagS. The third term in equation (13) takes into account coagulation between the particle in the diameter range $[\mathrm{dp}_i, \mathrm{dp}_{i+n}]$, called self-coagulation. Equation (13) does not include the flux of particles into, nor the flux of particles out from, the size range $[\mathrm{dp}_i, \mathrm{dp}_{i+n}]$ due to particle self-coagulation. Another, rather minor approximation made in equation (13) is that the coagulation terms are calculated at the lower boundary of each size bin.

For sizes at which atmospheric particle formation rates are usually determined (<10–20 nm diameter), dry deposition of particles is usually negligible compared with the particle removal rate by coagulation scavenging (Kerminen *et al* 2004). By neglecting the last term in equation (13) and rearranging, we obtain:

$$J_{\mathrm{dp}_i} \approx \frac{\Delta N}{\Delta t} + \frac{1}{2}\sum_{j=i}^{i+n-1} K(\mathrm{dp}_j, \mathrm{dp}_j) N_j^2 + \sum_{j=i}^{i+n-1} \mathrm{CoagS} \times (\mathrm{dp}_j)_{>\mathrm{dp}_{i+1}} N_j + J_{\mathrm{dp}_{i+n}} - \sum_{j=i}^{i+n-1} P_j. \tag{14}$$

While the approximations made above are not always valid, equation (14) can be considered as a general expression for the particle formation rate at the diameter dp_i in the atmosphere. Of the five terms on the right hand side of equation (14), the first one is obtained directly from the size distribution measurements, the second term can be calculated from the same measurements, and so can the third term when $\mathrm{dp}_{\max}$ is large enough so that the measured size range includes the dominant fraction of the CoagS (see equation (4)). The fourth and last terms are the trickiest ones and require a special attention in data analyses. Below we discuss shortly how equation (14) has commonly been used, and how it should be used, when analyzing atmospheric NPF events.

One way to use equation (14) is to consider a relatively narrow size range $[\mathrm{dp}_i, \mathrm{dp}_{i+n}]$, with dp_{i+n} well below 10 nm. The main advantage of this approach is that the last term of equation (14) representing primary particle emissions can usually be considered to be negligible. The drawback is that the flux of particles growing out of the size range $[\mathrm{dp}_i, \mathrm{dp}_{i+n}]$, i.e. the fourth term on the right hand side of equation (14), needs to be taken into account. Determining this flux requires making approximations on the particle number size distribution and GR at dp_{i+n} (see equation (5)), which causes potentially large errors in the calculated particle formation rate at dp_i (Vuollekoski *et al* 2012). Another way to use equation (14) is to consider a broader size range $[\mathrm{dp}_i, \mathrm{dp}_{i+n}]$, with dp_{i+n} being in the range 20–30 nm. In this case, neglecting the fourth term on the right hand side of equation (14) is usually a reasonable approximation. A potential problem, however, are primary particle emissions:

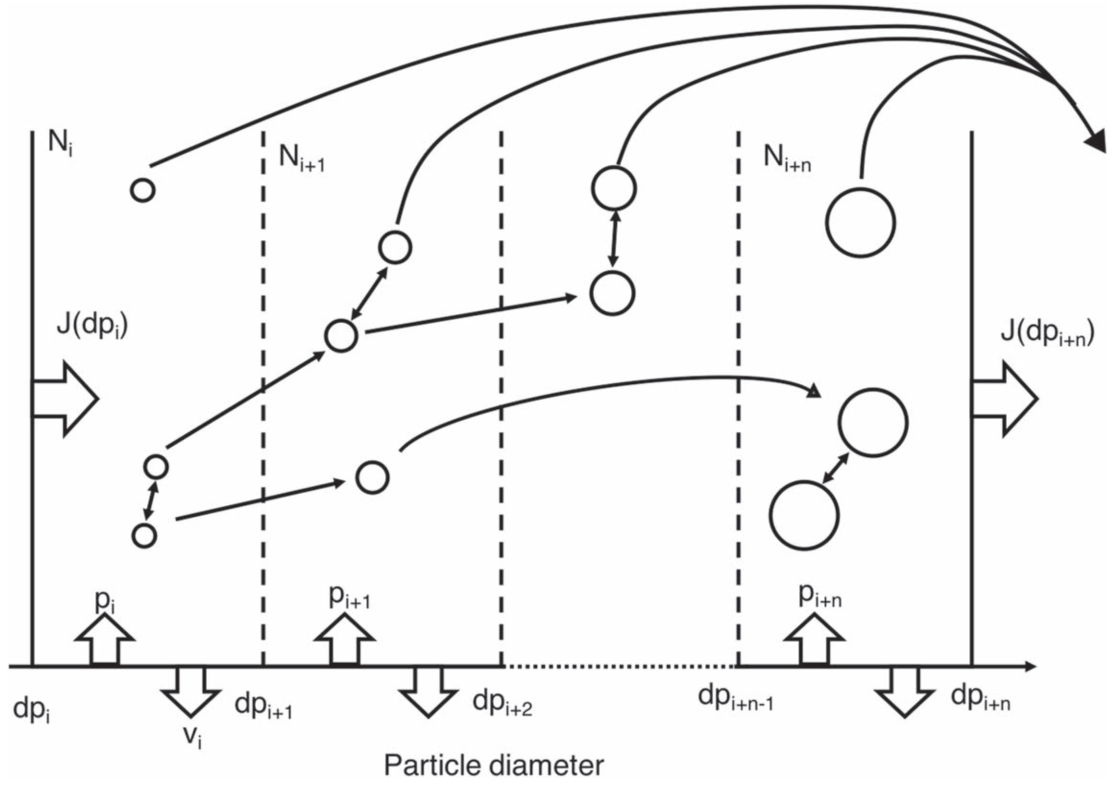

Figure 3. Illustration of the various terms in the balance equation of the particle number concentration in the size range [dp$_i$, dp$_{i+n}$] (see the text). One-headed arrows represent coagulation scavenging of particles, and the sum of the frequencies at which a particle in a certain size bin collides with all larger-size particles is equal to the coagulation sink of those particles. Two-headed arrows illustrate particle self-coagulation within each size bin, the corresponding coagulation coefficient being equal to K.

especially in urban environments, vehicular exhaust may be a substantial source of sub-20–30 nm particles (Ban-Weiss *et al* 2010, Brines *et al* 2015, Paasonen *et al* 2016), and these particles should be included in the fourth term of equation (14).

An issue worth noting is how the second and third terms on the right hand side of equation (14), representing coagulation losses, should be treated. Firstly, it is important to make a difference between these two terms, since for self-coagulation (the second term) the resulting aggregate will usually remain in the size range [dp$_i$, dp$_{i+n}$] while for coagulation with larger particles (the third term) it will not. Secondly, due to the relatively strong dependence of the CoagS on the particle size (see equation (6)), the third term on the right-hand side of equation (14) should be calculated separately for all the size bins belonging to the size range [dp$_i$, dp$_{i+n}$], rather than using a single 'representative' diameter in this size range. Accurate determination of the coagulation terms in equation (14) is especially important in highly-polluted environments where these terms may give a significant contribution to $\Delta N/\Delta t$ (Cai and Jiang 2017).

2.3.2. Particle GR

The particle GR can be determined from particle number size distribution measurements either with (e.g. Kuang *et al* 2012, Pichelstorfer *et al* 2018) or without (e.g. Kulmala *et al* 2012) considering the general dynamic equation for the growing particle population. The latter approach relies on two fundamentally different methods. In the first of these, termed the mode-fitting method (Dal Maso *et al* 2005, Kulmala *et al* 2012), a log-normal distribution function is fitted to a growing particle population, and some particle diameter characterizing this mode (number median diameter, mode peak diameter) is then determined. This procedure is repeated for successive time steps determined by the temporal resolution of the measurements, resulting in a series of particle diameter-time pairs (d_i, t_i; $i = 1 \ldots n$). In the second method, termed the appearance time method (Lehtipalo *et al* 2014), a series of particle diameter-time pairs is determined as well, but with a different logic: d_i is chosen to be some characteristic diameter of each measured size bin, while t_i is the time when the measured particle number concentration in this size bin reaches some pre-defined limit, typically its overall maximum or some fraction of that maximum.

In both the mode-fitting and appearance time method, the particle growth rate, GR, is obtained from the slope of a curve fitted to the obtained series of the particle diameter-time pairs (d_i, t_i; $i = 1 \ldots n$). This procedure provides typically a single value, and at most a few values, of GR for each NPF event. The resulting GR can therefore be interpreted as NPF event-average particle GR over a given particle diameter range. It should be noted that the size range, of which the obtained values of GR are representative, may vary considerably between different studies as well as between individual NPF events in a single study.

Estimating how the particle GR varies with time and particle size during a single NPF event involves large uncertainties, and is usually not possible without solving the general dynamic equation in combination with measurement data. The general principles of such approach, along with some examples on its application for measurement data, have been discussed in detail by Verheggen and Mozurkewich (2006), Kuang *et al* (2012), Yu *et al* (2016) and Pichelstorfer *et al* (2018), and thus will not be repeated here.

There is no method for the treatment of measurement data that can accurately reproduce the flux-equivalent particle GR defined by equation (5). For example, it has been shown using numerical simulations that the appearance time method discussed above systematically gives values of GR that exceed the corresponding flux-equivalent particle GR, and that this difference increases rapidly when going to sub-3 nm particle sizes (Olenius *et al* 2014, Kontkanen *et al* 2016b).

2.3.3. Formation and GRs of charged particles

There are several field measurement sites, for which no total particle number size distribution data are available, yet corresponding data for the charged fraction of the particle population exist. From such data, one can determine the formation rate of charged particles by using an equation that is otherwise similar to equation (14) but has two additional terms: a term that takes into account the loss of charged particles by ion–ion recombination, and a term that takes into account the attachment of small ions to neutral (uncharged) aerosol particles (see Manninen *et al* 2010, Kulmala *et al* 2012). Because of these two additional processes, the formation rate of charge particles is sensitive not only to the formation rate of charged clusters but also to the ambient ion concentration, the particle GR, and the size at which the formation rate is being determined (Kerminen *et al* 2007). As a result, one should be extremely careful in interpreting measurement data on the formation rate of charged particles, especially for sizes larger than about 2−3 nm in particle diameter, and in comparing such data between different environmental conditions.

Determining particle GRs from measured size distributions of charged particles can be done using the same procedures as outlined in section 2.3.2 for measured size distributions of total particle populations. When doing this, however, one needs to remember that the growth of a charged particle population differs somewhat from that of the total particle population. First, the condensation growth of charged aerosol particles is expected to be enhanced compared with similar-size neutral particles (Yu and Turco 2000, Nadykto and Yu 2003, Lushnikov and Kulmala 2004). Existing evidence suggest, however, that this effect is important only to the very smallest aerosol particles, being probably negligible for particles larger than 3 nm in diameter (e.g. Lehtipalo *et al* 2016). Second, coagulation losses of charged particles may be up to a factor of two higher than corresponding losses of neutral particles (Leppä *et al* 2011). This feature may affect calculated particle GRs in cases where coagulation losses give a significant contribution to the apparent particle GR (see equation (2)). Finally, the GR of charged particles might be affected by ion–ion recombination and ion-aerosol attachment (Gonser *et al* 2014). Only few field studies have compared the observed GRs of charged particles to those of the total particle population (Yli-Juuti *et al* 2009, Vakkari *et al* 2011, Yli-Juuti *et al* 2011, Hirsikko *et al* 2012, Gonser *et al* 2014). The overall message from these studies is that these two GRs typically compare relatively well with each other for particles larger than a few nm in diameter, while larger differences are occasionally seen for smaller particles. It is unclear to which extent these differences reflect real aerosol dynamical differences between the charged and neutral particles, and to which extent they arise from possible biases associated with different measurements or GR analysis methods.

3. Regional NPF

3.1. General character of regional NPF

3.1.1. Factors influencing the occurrence of NPF

The intensity of solar radiation reaching the Earth's surface is perhaps the most important factor in determining whether atmospheric NPF takes place or not. In practically all the measurement sites from where at least a few months of measurement data are available, the average solar radiation intensity was found to be higher during the NPF event days compared with non-event days (Birmili and Wiedensohler 2000, Vehkamäki *et al* 2004, Hamed *et al* 2007, Kristensson *et al* 2008, Jeong *et al* 2010, Guo *et al* 2012, Hirsikko *et al* 2012, Jun *et al* 2014, Kanawade *et al* 2014, Pierce *et al* 2014, Qi *et al* 2015, Wonaschütz *et al* 2015). No clear radiation threshold needed to initiate a regional NPF event has been identified, except at one study site (Lee *et al* 2008). The presence of clouds, by attenuating the solar radiation intensity below the cloud layer, decreases the probability of NPF to occur (Baranizadeh *et al* 2014, Dada *et al* 2017). An ongoing NPF event can be interrupted by the appearance of clouds (Hirsikko *et al* 2013), or by the occurrence of a solar eclipse (Jokinen *et al* 2017).

By acting as a sink for LVOC vapors and small clusters, a higher pre-existing aerosol loading is expected to hinder the occurrence of NPF. Existing observations confirm this expectation, since at most measurement sites the average value of CS was found to be lower on NPF event days compared with non-event days (Birmili *et al* 2003, Dal Maso *et al* 2007, Wu *et al* 2007, Asmi *et al* 2011, Pikridas *et al* 2012, Wang *et al* 2013, Young *et al* 2013, Kanawade *et al* 2014, Qi *et al* 2015, Salma *et al* 2016a, Dada *et al* 2017, Dai *et al* 2017). An exception to this pattern was the study by Birmili and Wiedensohler (2000) made in Melpitz, Germany, where the average pre-existing particle surface areas was somewhat higher on NPF event days compared with all the other days. In a couple urban locations, a threshold for CS, or aerosol surface area, above which NPF is very unlikely, was identified (e.g. Salma *et al* 2016a, Cai *et al* 2017). However, occurrences of NPF at very high values of CS have been observed (Nie *et al* 2014, Xiao *et al* 2015, Kulmala *et al* 2017).

The average ambient relative humidity (RH) tends to be lower on NPF event days than on non-event days in both clean and polluted environments (Birmili and Wiedensohler 2000, Birmili *et al* 2003, Vehkamäki *et al* 2004, Hamed *et al* 2007, Wu *et al* 2007, Lee *et al* 2008, Suni *et al* 2009, Guo *et al* 2012, Jun *et al* 2014, Kanawade *et al* 2014, Pierce *et al* 2014, Qi *et al* 2015, Zhao *et al* 2015, Salma *et al* 2016a, Dada *et al* 2017). Several possible reasons for this apparently close connection between the ambient RH and occurrence of NPF have been proposed, including the typically negative feedback of high RH on the solar radiation intensity, photochemical reactions and atmospheric lifetime of aerosol precursor vapors (e.g. Hamed *et al* 2011). In reality the situation is likely to be more complicated than this, since in many locations also air masses originating from very different source areas tend to be characterized by different levels of RH (e.g. Birmili *et al* 2003, O'Halloran *et al* 2009, Suni *et al* 2009, Dada *et al* 2017).

The effect of the ambient temperature (T) on NPF is ambiguous, showing very different responses between different studies. This feature is probably related to the simultaneous presence of several temperature-dependent processes that may either enhance or suppress NPF. Such processes include biogenic emissions of aerosol precursor vapors into the atmosphere and their oxidation to low-volatility vapors (e.g. Grote and Niinemets 2008), accumulation of aerosol particles which increases CS (e.g. Paasonen *et al* 2013), formation of molecular clusters and nanoparticles from various precursor vapors (e.g. Dunne *et al* 2016, Kürten *et al* 2016), and the diurnal evolution of the BL. Further complications arise from the strong seasonal cycle of the ambient temperature in many continental locations. For example, Dada *et al* (2017) found that in Hyytiälä, Finland, NPF is more frequent at higher values of T during the cold part of the year, while the opposite is true during the warm part of the year.

A close connection between the formation rate of new atmospheric aerosol particles and gas-phase sulfuric acid (H_2SO_4) concentration has been reported for a number of measurement sites (Weber *et al* 1995, 1996, 1997, Birmili *et al* 2003, Kulmala *et al* 2006a, Sihto *et al* 2006, Riipinen *et al* 2007, Kuang *et al* 2008, Nieminen *et al* 2009, Petäjä *et al* 2009, Paasonen *et al* 2010, Wang *et al* 2011, Yao *et al* 2018). Much less information is, however, available on how H_2SO_4 affects the occurrence of NPF, and even fewer studies have reported higher measured H_2SO_4 concentrations on NPF event days than on non-event days (Birmili *et al* 2003, Boy *et al* 2008, Wang *et al* 2011). Several studies have attempted to look at the relation between NPF and H_2SO_4 concentration by using some proxy variable for the gas phase H_2SO_4 concentration. Such analyses are subject to large uncertainties due to the scarcity of studies investigating the validity of these proxies in different environments (Petäjä *et al* 2009, Mikkonen *et al* 2011), and therefore will not be discussed here in more detail.

The main gaseous precursor for H_2SO_4 is sulfur dioxide (SO_2). Being also a major atmospheric pollutant that has undergone strict air quality regulations over the years, the potential connection between atmospheric NPF and SO_2 concentration has been of great interest. Observations in this regard are inconclusive: NPF event days have been reported to have both higher (Birmili and Wiedensohler 2000, Woo *et al* 2001, Dunn *et al* 2004, Boy *et al* 2008, Young *et al* 2013, Zhao *et al* 2015) and lower (Wu *et al* 2007, Dai *et al* 2017) ambient SO_2 concentrations, as compared with SO_2 concentrations measured during the non-event days or outside the periods of active NPF. In one study, the relation between the occurrence NPF and SO_2 concentration was found to be inconsistent between different seasons (Qi *et al* 2015). These contrasting observations can be understood by the delicate balance between the factors that favor (a higher gas-phase H_2SO_4 production rate) and disfavor (a larger sink for LVOC vapor and molecular clusters) NPF downwind major SO_2 sources. The potential influence of SO_2 emission reductions, driven mostly by past air quality regulations in United States and Europe, on the NPF event frequency will be discussed shortly in section 3.1.2.

Organic vapors, especially highly-oxygenated and extremely low-volatility organic compounds (ELVOC), have been speculated to participate actively into atmospheric NPF (e.g. Kulmala *et al* 1998, 2013, 2014, Ehn *et al* 2014, Jokinen *et al* 2015, Bianchi *et al* 2016). Since long-term ELVOC measurements are lacking at the moment, little can be said about how these compounds influence the occurrence of atmospheric NPF events. Dada *et al* (2017) showed, using 20 years of measurement data from Hyytiälä, Finland, that a proxy for the concentration of oxidized organic compounds in the gas phase (see section 2.2.2) was, on average, higher on NPF event than on non-event days in every month of the year. This observation gives some confidence that the ELVOC concentration might be among the most important variables affecting occurrence of NPF in the continental atmosphere.

In a number of studies, the probability by which NPF events occur has been found to depend strongly on the wind direction or, more specifically, on the origin of measured air masses (e.g. Hamed *et al* 2007, Sogacheva *et al* 2007, Suni *et al* 2009, Asmi *et al* 2011, Shen *et al* 2011, Vakkari *et al* 2011, Nieminen *et al* 2014, Qi *et al* 2015, Mordas *et al* 2016, Kolesar *et al* 2017). Such dependency is quite expected, since in most locations air masses coming from different directions tend to be affected by different levels of biogenic emissions and anthropogenic pollutants, and exposed to different meteorological conditions prior to their arrival at the measurement site.

The above discussion summarizes our current understanding on how different atmospheric variables are related to the occurrence of NPF. Over the years, people have also searched for more general criteria to predict whether NPF takes place or not under given atmospheric conditions. Based on the pioneering theoretical work by McMurry and Friedlander (1979) and McMurry (1983), McMurry *et al* (2005) developed a parameter 'L' to distinguish the days with NPF from those without NPF in a sulfur-rich environment. Kuang *et al* (2010) extended this work to more diverse environments with a parameter 'L_Γ' and demonstrated the value of this parameter to be a good predictor for the occurrence of NPF. Kulmala *et al* (2017) introduced a dimensionless survival parameter 'P', a variable closely related to L_Γ, and pointed out that we are still lacking a general understanding on why NPF occurs under extremely-polluted conditions. The main problem in applying L, L_Γ or P to predicting the occurrence of NPF is that determining their values requires knowing either the cluster GR or gas-phase concentration of vapors causing this growth. Such information is rarely available from atmospheric measurements. By relying on some combination of variables known to either favor or disfavor NPF, people have developed such NPF predictors that can relatively easily be derived from routine measurement data (e.g. Clement *et al* 2001, Boy and Kulmala 2002, Hyvönen *et al* 2005, Mikkonen *et al* 2006, Jayaratne *et al* 2015, Nieminen *et al* 2015). While many of these NPF predictors seem to work well for limited data sets, or constrained atmospheric conditions, none of them has been shown to have a universal predictive power in the continental troposphere.

3.1.2. Temporal characteristics

Regional NPF is typically a daytime phenomenon. The studies covering at least two full years of measurements have reported that the starting times of NPF formation events are confined almost exclusively between the sunrise and sunset (Vehkamäki *et al* 2004, Hamed *et al* 2007, Wu *et al* 2007, Asmi *et al* 2011, Qi *et al* 2015). Furthermore, the active period of NPF tends to end before sunset (Hamed *et al* 2007, Wu *et al* 2007, Qi *et al* 2015). Nighttime NPF have been reported in a few locations, including a boreal forest and its surroundings (Vehkamäki *et al* 2004, Junninen *et al* 2008, Svenningsson *et al* 2008, Buenrostro Mazon *et al* 2016, Rose *et al* 2018), a pine forest in France (Kammer *et al* 2018), an Eucalypt forest in Australia (Suni *et al* 2008), a Mediterranean Island (Kalivitis *et al* 2012), a rural site affected by orographic cloud processing (Wiedensohler *et al* 1997), an industrial complex in South Africa (Hirsikko *et al* 2012), and an urban site in Australia (Salimi *et al* 2017, Pushpawela *et al* 2018). In some of these locations the particle growth following nighttime NPF was very limited (e.g. Kalivitis *et al* 2012, Buenrostro Mazon *et al* 2016, Rose *et al* 2018), while in some locations such growth could be very intense, producing particles of several tens of nm in diameter (Suni *et al* 2008, Svenningsson *et al* 2008). Two aircraft studies have reported indications of nighttime NPF in the free troposphere (FT) (Lee *et al* 2008, Rose *et al* 2015a). All these features, together with findings related to the solar radiation intensity (see section 3.1.1), tend to imply a very crucial role of atmospheric photochemistry in maintaining regional NPF and growth.

The duration of a NPF event refers to the length of the period during which active NPF is taking place or, in a more practical sense, the length of the period during which very small particles enter the measured particle size range. Very few publications report the duration of the observed NPF events, even though such information is essential when determining the event-average NPF rates.

A typical feature of regional NPF events is that the newly-formed particles grow gradually into larger sizes. In theory, this growth continues until the particles are removed from the atmosphere by deposition or coagulation processes. In practice, however, one can track the growth of newly-formed particles from a few hours up to a day or two when using measurements conducted at a fixed location. Several factors contribute to our inability to follow the particle growth further in time, including the diurnal evolution of the continental BL and associated mixing, limited spatial extent of NPF (see section 2.3.3), and general difficulties in separating 'aged' particles of different origin using routine measurements. In a vast majority of cases, the growth of newly-formed particles appears to be irreversible. However, there are a few locations where the newly-formed particles first grow in size for a few hours and then, occasionally, appear to shrink to various extents (Yao *et al* 2010, Cusack *et al* 2013, Young *et al* 2013, Skrabalova *et al* 2015, Lihavainen *et al* 2016, Salma *et al* 2016b, Zhang *et al* 2016, Alonso-Blanco *et al* 2017). For a more detailed discussion on this phenomenon, we refer to a recent paper by Alonso-Blanco *et al* (2017).

In a few locations, the occurrence of two or more NPF events during the same day have been reported (Suni *et al* 2008, Svenningsson *et al* 2008, Hirsikko *et al* 2013, Kyrö *et al* 2013, Rose *et al* 2015b, Salma *et al* 2016a). Such phenomenon, while rather rare, has several possible causes. First, the presence of clouds can interrupt an ongoing NPF event for a while, after which either the same event will continue or a new event will begin (e.g. Hirsikko *et al* 2013). Second, it is possible that due to changing air masses or evolving chemical composition in the same air mass being measured, two different types of regional NPF events are being initiated at different times of the same day (e.g. Salma *et al* 2016a). Finally, sometimes a 'local' NPF event may emerge on top of a regional NPF event (e.g. Kyrö *et al* 2013).

The frequency of NPF occurrence varies over the course of a year, since most of the variables influencing NPF have a pronounced seasonal variation. In a vast majority of the sites, NPF is more frequent in summer compared with winter (Qian *et al* 2007, Dall'Osto *et al* 2018, Nieminen *et al* 2018). The season with the highest NPF event frequency tends to change from summer in polar and many high-latitude regions (Heintzenberg *et al* 2017, Nieminen *et al* 2018) toward the spring or autumn in most other regions (e.g. Jeong *et al* 2004, Stanier *et al* 2004, Dal Maso *et al* 2005, Hussein *et al* 2008, Pryor *et al* 2010, Asmi *et al* 2011, Kyrö *et al* 2014, Asmi *et al* 2016, Mahish and Collins 2017, Wang *et al* 2017b, Nieminen *et al* 2018). This pattern is, however, by no means universal, and the overall ranking order of the seasons in terms of their NPF frequency shows a substantial variability between individual measurements sites (e.g Wu *et al* 2007, Meija and Morawska 2009, Manninen *et al* 2010, Shen *et al* 2011, Vakkari *et al* 2011, Hirsikko *et al* 2012, Qi *et al* 2015, Wonaschütz *et al* 2015, Dall'Osto *et al* 2018). Very few studies have found NPF to be most frequent during the wintertime (Lee *et al* 2008, Pikridas *et al* 2012). Although the NPF frequency has varying seasonal characteristics at different sites, the GRs of newly-formed particle display almost exclusive a summer maximum (e.g. Nieminen *et al* 2018). This feature originates from higher biogenic emissions and typically stronger atmospheric photochemistry during the summertime, both of them enhancing the production of LVOC vapors responsible for the particle growth, while there is practically nothing during summer (except perhaps extreme temperatures) that would be expected to suppress the particle growth.

Changes in anthropogenic emissions, along with climate change, are expected to affect the probability of occurrence of atmospheric NPF. Unfortunately, very few observational data on long-term changes in the NPF event frequency exist. Hamed *et al* (2010) showed that both the intensity and frequency of NPF decreased considerably between two time periods (1996−1997; 2003−2006) at a rural site in Melpitz, Germany, and hypothesized that this decrease was mainly due to the major decline in gas-phase SO_2 concentrations over the same time period. Wang *et al* (2017b) demonstrated a further decline in NPF and SO_2 concentrations from 2003 to 2011, so that that annual NPF event frequency decreased from 40%−50% in 1996−1997 down to <20% during 2007−2011 in Melpitz. Saha *et al* (2018) reported that the regional NPF event frequency decreased from about 30% during 2001−2002 to about 10% during 2016−2017 In Pittsburg, Pensylvania, in United States, accompanied by a strong decline in the SO_2 concentration of about 90% between these two time periods. Kyrö *et al* (2014) observed a strong decline ($-3.7\%\ \mathrm{yr}^{-1}$) in the NPF event frequency at Värriö in Northern Finland over the time period of 1998−2011, and attributed this decline to decreasing sulfur emissions from the nearby industrial sources in the Kola Peninsula. Pallas, a site about 300 km west from Värriö, however, displayed no trend in the NPF event frequency during 2000−2010 (Asmi *et al* 2011). The study by Asmi *et al* (2011) did not report the temporal behavior of the SO_2 concentration in Pallas, but it is known that compared with Värriö, Pallas is considerably less affected by SO_2 emissions from the Kola Peninsula. In Hyytiälä, Finland, particle formation and GRs show a slight positive trend ($0.2\%-0.5\%\ \mathrm{yr}^{-1}$) since 1996 (Nieminen *et al* 2014), whereas no clear trend but a prominent inter-annual variability can be seen for the NPF event frequency (Nieminen *et al* 2014, Dada *et al* 2017). Finally, Kalivitis *et al* (2018) reported a slight increase in the NPF event frequency over the period 2008–2015 at Crete, Greece, in the eastern Mediterranean.

3.1.3. Spatial characteristics

Several studies have attempted to estimate the spatial extent of regional NPF. For this purpose, various methods based on single-site measurements have been developed (e.g. Birmili *et al* 2003, Hussein *et al* 2009, Crippa and Pryor 2013, Kristensson *et al* 2014, Nemeth and Salma 2014). A more detailed view on the spatial extent and variability of NPF can be obtained by using simultaneous measurements from two or more stations, and such analyses have been performed for Northern (Tunved *et al* 2003, Vana *et al* 2004, Komppula *et al* 2006, Hussein *et al* 2009) and Central Europe (Wehner *et al* 2007), the Carpathian Basin (Salma *et al* 2016a), the Mediterranean atmosphere (Berland *et al* 2017), Eastern North America (Crippa and Pryor 2013), Ontario, Canada (Jeong *et al* 2010, Jun *et al* 2014), the Korean Peninsula (Kim *et al* 2016), North China Plain (Wang *et al* 2013) and Eastern China (Shen *et al* 2018). The general conclusion from these studies is that the spatial extent of regional NPF is typically a few hundreds of km, and possibly exceeding 1000 km in some environments.

Despite the usually relatively large spatial extent of regional NPF, most of the studies mentioned above observed a notable variability in the timing, duration and intensity of NPF events across the study region. This spatial variability appears to be larger for particle formation rates compared with particle GRs. Paired

Table 1. Statistics (median, 5th and 95th percentiles) of the particle formation rates (J) and growth rates (GR) based on literature data. Mountain sites include studies conducted in China. For each site type, N refers to the number of sites from which the median values of J and GR were determined. As an example, the median GR of 2.7 nm h^{-1} for boreal forest sites is the median value of the 17 study-average (or median if mean was not reported) values of GR reported in each individual study. It should be noted that the size range used in calculating J and GR varied from study to another (e.g. J could refer to J_3, J_{10} etc), and we had no way of harmonizing the results in this respect.

		J (cm^{-3} s^{-1})				GR (nm h^{-1})		
Site type	N	5th	Median	95th	N	5th	Median	95th
Boreal	12	0.13	0.4	0.92	17	0.49	2.7	5.3
Remote and rural	6	0.59	4.1	11.0	22	2.0	3.5	9.6
Urban	17	1.2	2.9	13.7	26	4.0	5.9	12
Arctic	2	—	0.51	—	6	0.23	2.3	4.1
Antarctica	2	—	0.05	—	4	1.4	4.5	5.5
Mountain	10	0.2	0.79	3.9	11	1.2	4.0	13
China: rural	11	1.8	4.9	19.7	13	3.8	6.2	9.8
China: suburban	4	1.4	3.3	3.6	9	3.5	7.4	13
China: urban	8	1.8	7.9	12.9	16	4.1	6.4	12
China: marine and coastal	1	—	0.3	—	5	2.9	4.5	7.1

urban and rural locations display interesting features in this regard: in comparison with rural sites, NPF in the nearby urban sites tend to be less frequent (Yue *et al* 2013, Jun *et al* 2014, Salma *et al* 2016a), yet more intense in terms of observed particle formation and GRs (Wang *et al* 2013, Salma *et al* 2016a). Furthermore, when occurring, regional NPF may start, on average, either later (Wehner *et al* 2007, Salma *et al* 2016a) or earlier (Jung *et al* 2013, Yue *et al* 2013) at the urban site than at the nearby rural site. Observations indicate that the spatial variability of regional NPF is apparent not only between the urban and rural locations, but also within an urban area (e.g. Siakavaras *et al* 2016) and over a rural or sea areas (e.g Crumeyrolle *et al* 2010, Crippa and Pryor 2013, Berland *et al* 2017). Challenges in distinguishing between regional and local NPF events at sub-regional scales makes it difficult to perform detailed analyses of small-scale variabilities in regional NPF.

The vertical extent of regional NPF has been investigated in rather few studies. Airborne observations suggest that in some regions NPF and subsequent particle growth seem to be confined into the BL (O'Dowd *et al* 2009, Crumeyrolle *et al* 2010), while in other regions this phenomenon may also take place in the FT (Rose *et al* 2015a, Bianchi *et al* 2016, Berland *et al* 2017) or at the interface between the BL and FT (Siebert *et al* 2004, Dadashazar *et al* 2018). In Beijing, China, the height in the atmosphere having the strongest NPF was observed to move from within the BL at low aerosol loadings to the top of the BL or to the lower FT at high aerosol loadings (Quan *et al* 2017). Observations in several mountain-top locations indicate a relatively frequent occurrence of seemingly regional NPF events at high altitudes (see section 3.2.5). It is unclear how big fraction of these NPF events is truly of free tropospheric origin, and whether they are related to a recent transport of aerosol precursors from the BL.

It would be tempting to conclude that the spatial extent and variability of regional NPF is affected solely by spatial inhomogeneities in the sources of aerosol precursor compounds. However, the delicate balance between the factors that favor atmospheric NPF on one hand, and the factors that suppress it on the other hand, changes this picture. Tunved *et al* (2006) showed that in originally clear air entering a boreal forest zone, atmospheric NPF is first initiated after the air mass have traveled some time over the forested area, and then maintained until the increasing pre-existing aerosol loading kills it. In this case, the spatial extent of NPF as well as its temporal characteristics at any fixed location are dictated by the rate at which the transported air masses accumulate aerosol precursor compounds. Another example of the dynamic nature of regional NPF comes from Beijing, China, where the occurrence of NPF was found to be tied to a multi-day cycle of air pollutant accumulation (Guo *et al* 2014). These examples demonstrate the complex interplay between the spatial extent and temporal characteristics of regional NPF—a feature that is rarely taken into account when analyzing NPF observations made at fixed locations.

3.2. Observed particle formation and GRs and NPF event frequencies

In this section we summarize the literature results on reported intensities and frequencies of regional NPF in the following types of environments: rural and remote continental areas, urban environments, Arctic region and Antarctica, marine areas, as well as FT and mountain sites. Table 1 summarizes the main features of observed particle formation and GRs in these environments, while more detailed information on the reported character of NPF in each individual study and location can be found in the supplementary tables is available online at stacks.iop.org/ERL/13/103003/mmedia. Figure 4 displays the geographic location of

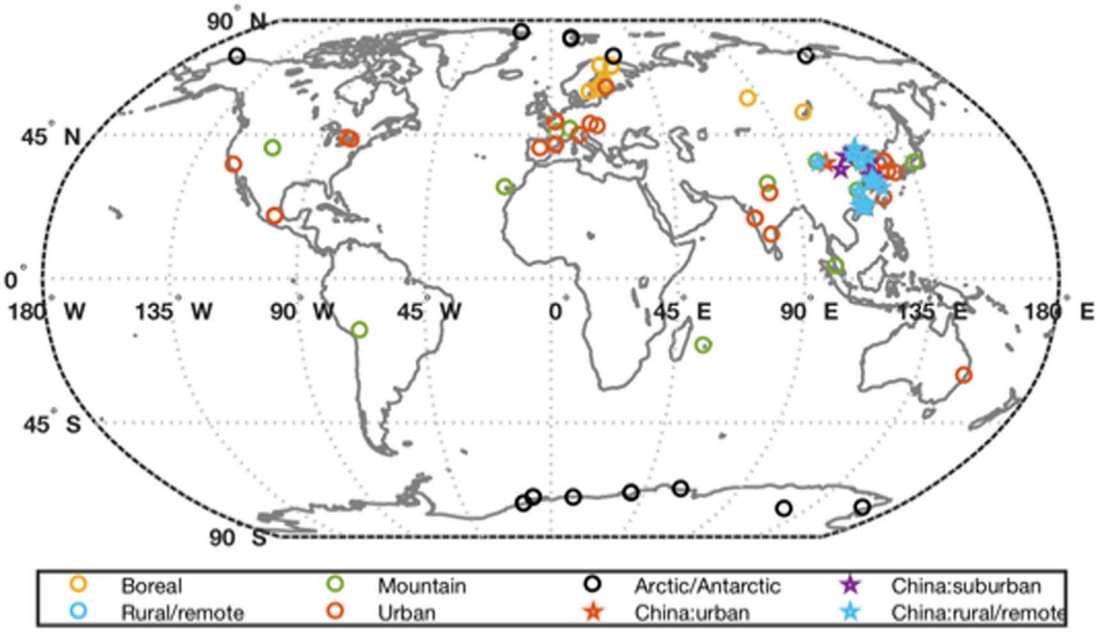

Figure 4. Geographical locations of measurement sites in different environments. The map contains 183 sites.This plot is made from all available coordinates reported in the literature collected in the supplementary tables. The Chinese sites are collected from studies conducted in the People's Republic of China.

all the measurement sites included in the supplementary tables and in our analysis.

3.2.1. Rural and remote continental areas

Published analyses of regional NPF events in rural and remote continental areas cover a wide range of environments, including boreal and other forested areas, agricultural regions, grasslands and various types of environments located at different distances from major urban centers. In the following we consider boreal forest sites separately from other types of sites, mainly because the boreal forests are the only environment having long-term measurements of atmospheric NPF from several different sites. Arctic and polar continental environments, as well as mountains sites, will be considered separately in sections 3.2.3 and 3.2.5.

The SMEAR II station (Hari and Kulmala 2005), located in a boreal forest environment in Hyytiälä, Finland, was the first field site from where regional NPF events were reported in a scientific literature (Mäkelä *et al* 1997), and the same site has so far the longest published times series of such events (Nieminen *et al* 2014). The observed particle formation and GRs span roughly an order of magnitude between the different boreal forest sites, the median values being 0.4 cm^{-3} s^{-1} and 2.7 nm h^{-1}, respectively (table 1). The annual frequency of NPF event varies between about 10% and 30%, with lowest frequencies corresponding to the northern edge of the boreal forest zone and the Siberian part of this region. The NPF event frequency has often a maximum in spring, possibly another maximum in late summer or autumn, and is typically very low during the wintertime (Dal Maso *et al* 2007, 2008, Kristensson *et al* 2008, Asmi *et al* 2011, Nieminen *et al* 2014, Kyrö *et al* 2014).

Particle formation rates in rural and remote locations other than boreal forest sites span over a relatively large range of values (table 1), as one might expect due to the variable characteristics of these environments. The median value of J from these environments is about ten times higher than that from the boreal forest sites, and the same concerns the high- and low-end values of this quantity between the rural China and boreal forest environment. Compared with J, smaller differences exist in GR between the rural and remote locations, and the median value of this quantity outside China is only slightly higher than that in boreal forests. Typical particle GRs in Chinese rural sites are about twice those in rural sites elsewhere in the world.

The frequency of regional NPF in rural and remote locations varies a lot. At one extreme, NPF event frequencies between about 70% and 90% were reported in South Africa (Vakkari *et al* 2011, Hirsikko *et al* 2012, Vakkari *et al* 2015). At the other extreme, NPF was found to be quite rare around a forested location in Siberia (Heintzenberg *et al* 2011), and almost nonexistent in the Amazonian rainforest (Martin *et al* 2010, Wimmer *et al* 2018, Andreae *et al* 2018, Rizzo *et al* 2018).

3.2.2. Urban environments

Since the last review that concentrated on observed atmospheric NPF (Kulmala *et al* 2004), a large number of studies have investigated NPF in various types of urban environments. The median particle formation rates are the lowest in Antarctica, followed Arctic and boreal forest sites, and still higher in other remote and rural environments as well as in urban areas (table 1). In China, high particle formation rates tend to be observed in almost all the sites.

The median particle GR from the urban sites is 1.5–2 times that from the non-Chinese rural and remote locations (table 1). Somewhat larger differences can be seen at the low-end values of this quantity, mainly because observed particle GRs smaller than about 1 nm h^{-1} are rare in urban environments. It could be mentioned that particle GRs >10 nm h^{-1} are common in many urban locations, especially in China, but seemingly also in some other polluted environments (Mönkkönen *et al* 2005, Wu *et al* 2007, Iida *et al* 2008, Kalafut-Pettibone *et al* 2011, Peng *et al* 2014, Qi *et al* 2015, Xiao *et al* 2015, Zhao *et al* 2015).

Typical frequencies of regional NPF events in urban environments are in the range of 10%–30% (Stanier *et al* 2004, Qian *et al* 2007, Dall'Osto *et al* 2013, Brines *et al* 2015, Wonaschütz *et al* 2015, Hofman *et al* 2016, Salma *et al* 2016a). Higher NPF event frequencies were observed in Po Valley, Italy (36%, Hamed *et al* 2007), London, United Kingdom (36%, Hofman *et al* 2016), Beijing, China (40%, Wu *et al* 2007), and Nanjing, China (44%, Qi *et al* 2015). NPF event frequencies as low as 5% were reported in Birmingham, United Kingdom (Alam *et al* 2003) and in Helsinki, Finland (Hussein *et al* 2008). No consistent pattern in the seasonal variability of the NPF event frequency in urban areas can be identified.

3.2.3. Arctic areas and Antarctica

Particle number concentration and size distribution measurements conducted in the Arctic suggest that this region is an active area of NPF during the summertime (Leaitch *et al* 2013, Tunved *et al* 2013, Freud *et al* 2017). This view is supported by the few published regional NPF event frequencies, reaching values between about 30% and 40% in Tiksi, northern Russia, and in Station Nord, northeast Greenland (Asmi *et al* 2016, Nguyen *et al* 2016). Summertime NPF was found to be frequent also in air masses measured at the Zeppelin station, Svalbard (Dall'Osto *et al* 2017, Heintzenberg *et al* 2017), as well as in Canadian Arctic marine and coastal environments (Collings *et al* 2017). Interestingly, the summertime NPF event frequencies in the Arctic seem to be clearly higher than those reported at a few sub-Arctic sites at the edge of the boreal forest zone at the same time of the year. The frequency of NPF during the winter is very low in most Arctic locations. Dall'Osto *et al* (2017) reported an anti-correlation between the NPF event frequency in Svalbard and both monthly and annual extent of Arctic sea ice area, suggesting that future reductions in the Arctic sea ice extent might lead to enhanced NPF over the Arctic.

Although cases of regional NPF have been observed throughout the Arctic, data on NPF and GRs are available from few sites only (table 1). Based on these rather limited data, particle formation and GRs in the Arctic atmosphere appear to be comparable to those observed in boreal forest environments.

It should be noted that in addition to NPF taking place inside the Arctic BL, sources of nucleation and small Aitken mode particles might also be the entrainment from the FT (e.g. Tunved *et al* 2013, Croft *et al* 2016, Igel *et al* 2017, see also section 3.2.4), or primary particles emitted from the Arctic Ocean (Leck and Bigg 1999, 2005, Orellana *et al* 2011, Karl *et al* 2013). Heintzenberg *et al* (2017) applied three different types of search algorithms for NPF and particle growth events in the Svalbard region over a 10 year measurement period, partly reflecting the different sub-60 nm particle sources, and found evidence on varying and relatively complex marine biological source processes for these particles.

Over Antarctica, a prominent feature of the aerosol system is the very pronounced annual cycle of the total particle number concentration, being up to 20–100 times higher during the austral summer than during the winter (Shaw 1988, Gras 1993, Ito 1993, Hara *et al* 2011, Weller *et al* 2011, Järvinen *et al* 2013, Fiebig *et al* 2014, Kim *et al* 2017). The magnitude of this seasonal cycle appears to be higher on the upper plateau of Antarctica than at the coastal Antarctic sites, whereas overall particle number concentration levels are clearly higher at the coastal Antarctica. Particle number size distribution measurements suggest that the summer maximum in particle concentrations can, to a large extend, be explained by NPF taking place in the Antarctic atmosphere. The vertical location of Antarctic NPF has not been well quantified, even though there are some indications that NPF takes more preferably place in the Antarctic FT than in the BL (Koponen *et al* 2002, Hara *et al* 2011, Humphries *et al* 2016).

Regional NPF has been investigated mainly during the summertime in Antarctica, and there are too few data to estimate the frequency of occurrence of this phenomenon. The available information suggests, however, that regional NPF events observed at the surface level are probably less common in the summer Antarctica than they are in the summer Arctic atmosphere (Koponen *et al* 2003, Park *et al* 2004, Pant *et al* 2011, Järvinen *et al* 2013, Kyrö *et al* 2013, Weller *et al* 2015). A study conducted on the upper plateau of Antarctica demonstrates that also wintertime regional NPF is possible in this environment (Järvinen *et al* 2013). Typical particle formation rates associated with regional NPF in Antarctica are about an order of magnitude lower than those in the Arctic or boreal forest environments, whereas the corresponding particle GRs are comparable to those in other environments (see table 1). In the Antarctic atmosphere, also cases displaying very low particle GRs between about 0.1

and 1 nm h^{-1} were reported (e.g. Park *et al* 2004, Weller *et al* 2015).

3.2.4. Marine areas

When discussing NPF in marine areas, it is important to distinguish between remote marine areas with minimal anthropogenic influence, continental outflow regions with potentially large anthropogenic effects on NPF (e.g. McNaughton *et al* 2004), and coastal regions which may have their own NPF mechanisms (see section 4.2). Below we discuss shortly NPF taking place in the remote marine areas, with an emphasis on the marine boundary layer (MBL). This region has been of great research interest after the proposed feedback mechanisms between marine sulfur emissions and climate (Charlson *et al* 1987), in which increased ocean temperatures in a warming climate would cause larger dimethyl sulfide (DMS) emissions to the atmosphere and subsequently increased CCN production due to more efficient NPF and particle growth. Higher CCN concentrations would then result in higher albedos of MBL clouds, causing a cooling effect that would partly compensate for the initial climate warming. The various steps and overall importance of this climate feedback mechanism have been investigated, and debated, a lot during the past three decades (e.g. Ayers and Cainey 2007, Woodhouse *et al* 2010, Quinn and Bates 2011, Thomas *et al* 2011, Mahajan *et al* 2015).

Indications on NPF inside the MBL have been reported from several marine locations (e.g. Covert *et al* 1992, Hegg *et al* 1993, Clarke *et al* 1998a, Weber *et al* 1999, Petters *et al* 2006). However, excluding high-latitude marine areas (see section 3.2.3), this phenomenon appears to be sporadic in nature (Covert *et al* 1996, Heintzenberg *et al* 2004), requiring quite specific conditions to occur like a very low pre-existing aerosol loading. Regional NPF with subsequent particle growth to larger sizes have been observed only outside tropical MBL, and the reported particle GRs are relatively low, of the order of 1 nm h^{-1} (O'Dowd *et al* 2010, Ueda *et al* 2016).

Following the observations by Bigg *et al* (1984) and Clarke (1993), Raes (1995) suggested that, instead of NPF inside the MBL, entrainment of particles formed initially in the FT would be the main source of secondary aerosol particles in the remote MBL. Strong support for this hypothesis was later obtained from compilations of ship cruises and aircraft missions (Covert *et al* 1996, Clarke and Kapustin 2002), as well as from field campaigns combining different types of measurements (Raes *et al* 1997, Kamra *et al* 2003, Clarke *et al* 2013, Quinn *et al* 2017). The view that regional NPF does not commonly occur in mid- and low-latitude remote MBLs, and that both ultrafine particle and CCN populations in these regions are maintained by a combination of sea-spray emissions from the ocean surface and entrainment from the FT, is further supported by models simulating processes determining the MBL aerosol particle budget (Capaldo *et al* 1999, Katoshevski *et al* 1999, Pirjola *et al* 2000), as well as by large-scale model simulations (e.g. Korhonen *et al* 2008, Woodhouse *et al* 2010).

3.2.5. FT and mountain sites

The near absence of primary particle sources and longer aerosol particle lifetimes in the FT compared with the BL make this environment an interesting location from the atmospheric NPF point of view. Furthermore, as discussed is section 3.2.4, FT has been identified as a potentially important source of newly-formed particles into the BL. The characteristics of NPF in different regions and heights of the FT have been investigated with airborne measurements and by conducting measurements at fixed locations on mountains. Below we discuss shortly our current understanding on NPF in the FT based on these two experimental approaches.

By relying on high-altitude aircraft measurements in the FT, Clarke (1993) found an inverse relation between the sub-15 nm particle concentration and aerosol surface area, and concluded that the upper part of the FT provides conditions favorable for NPF. Later measurements confirmed the upper FT as an active area of NPF, especially in the tropics but also in the northern-hemispheric mid-latitudes (Zaizen *et al* 1996, de Reus *et al* 2001, Clarke and Kapustin 2002, Singh *et al* 2002, Heintzenberg *et al* 2003, Hermann *et al* 2003, Weigelt *et al* 2009, Takegawa *et al* 2014). Several studies reported associations between NPF in the upper FT and convective uplifting, possibly accompanied with emissions from continental surface sources (e.g. Wang *et al* 2000, Heintzenberg *et al* 2003, Benson *et al* 2008, Köppe *et al* 2009). In many studies, the outflow regions of convective clouds were identified as an active location of NPF (see section 4.3), but there is no proof that the presence of clouds would be a necessary condition for free-tropospheric NPF. In addition to convection, tropopause folds were observed to initiate NPF in the upper FT (Young *et al* 2007).

Measurements at mountain stations make it possible to investigate regional NPF in the FT, and its connection with FT-BL interactions. Continuous measurements at mountain sites reveal variable frequencies of NPF event days: 64% with a maximum close to 100% during the dry season at Chacaltaya (5240 m a.s.l) in Bolivia (Rose *et al* 2015b), >35% with a maximum close to 50% during the monsoon and post-monsoon seasons at Pyramid station (5079 m a.s.l) in Nepal (Venzac *et al* 2008), 15% with moderate spring and late summer maxima at Jungfraujoch (3580 m a.s.l) in Switzerland (Herrmann *et al* 2015), 52% with a spring maximum and summer minimum at Storm Peak Laboratory (3210 m a.s.l) in Colorado, United States (Hallar *et al* 2011, 2016), 30% with a summer maximum of about 50% at Izana (2373 m a.s.l) in Tenerife island (Garcia *et al* 2014), 65% with an autumn maximum of about 90% at Maido observatory (2150 m a.s.l) on Reunion Island (Foucart et al 2018), 11% with a spring maximum of about 20% at Mukteshwar (2180 m a.s.l) in India (Neitola *et al* 2011), 32% with a spring maximum of 75% at Mount Tai (1534 m a.s.l) in China (Shen *et al* 2016a), and 23% with no clear seasonal prefence at Puy de Dome (1465 m a.s.l) in central France (Rose *et al* 2013). Signs of frequent NPF were also reported from three other high-altitude sites: Mount Saraswati (4520 m a.s.l) in the Trans-Himalayan region (Krishna Moorthy *et al* 2011), Mount Waliguan (3816 m a.s.l) in China (Kivekäs *et al* 2009) and Mount Lemmon (2790 m a.s.l) in Arizona, United States (Shaw 2007).

In several mountain sites, NPF was found to be strongly associated with upslope valley winds bringing air from lower altitudes, plausibly from the BL (Weber *et al* 1995, Shaw 2007, Nishita *et al* 2008, Venzac *et al* 2008, Rodriqez *et al* 2009, Shen *et al* 2016a). In Mukteshwar, a low-altitude mountain site in India, NPF was common only during the spring months when the site was located within the BL (Neitola *et al* 2011). In Jungfraujoch, Switzerland, NPF was found to be restricted to air masses that had been in contact with the BL within the last couple of days (Bianchi *et al* 2016). These observations give further support for the important role of surface emissions in causing NPF in the FT.

Reported particle formation rates from different mountain sites span over a relatively large range of values, with the median value being slightly larger than that in the boreal forest environment yet considerably lower than that in other remote or rural environments (table 1). Reported particle GRs at mountain sites are relatively high, comparable to those in many of the sites in the continental BL.

4. NPF taking place at sub-regional scales

4.1. Plumes from ground-based point sources

Coal-fired power plant plumes were among the first places in the atmosphere where extensive NPF was observed to take place (e.g. Dittenhoefer and de Pena 1978, Whitby *et al* 1978, Hobbs *et al* 1979, Van Valin and Pueschel 1981, Wilson and McMurry 1981). The formation rates of new aerosol particles in these plumes were reported to reach values larger than 1000 cm^{-3} s^{-1}, and the particle production was typically found to be the strongest near plume edges. In many cases, signs of NPF could be observed at distances of several tens of km downwind from the source. Later studies reported active NPF in many other types of plumes originating from anthropogenic point sources. Such sources include various kinds of power plants, refineries, smelters and other industrial complexes (e.g. Brock *et al* 2003, Banic *et al* 2006, Junkermann *et al* 2011a, Junkermann and Hacker 2015).

Biomass burning is one of the largest sources of aerosol particles and many trace gases in the global atmosphere (Andreae and Merlet 2001, Van Marle *et al* 2017). Laboratory experiments conducted by Hennigan *et al* (2012) demonstrated that biomass burning plumes have a great potential for NPF and subsequent particle growth, yet there are very few field studies on this phenomenon. Indirect evidence of NPF has been reported in plumes originating from vastly different biomass burning sources (Hobbs *et al* 2003, Bougiatioti *et al* 2016, Laing *et al* 2016). By segregating a large number of savannah fire plumes of different age together, Vakkari *et al* (2014) observed intense NPF followed by a rapid growth of newly-formed particles in these plumes during daytime. The above studies indicate that, in addition to studying trace gas and primary particle emissions from biomass burning, emphasis should also be put on investigating NPF taking place in plumes from different biomass burning sources.

Several studies have reported rapid conversion of SO_2 to sulfate aerosol in power plant plumes (Hewitt 2001), as a result of which existing parameterizations on NPF associated with power plant and other sulfur-rich plumes tend to rely heavily on the amount of SO_2 emitted by these plumes (see Stevens and Pierce, 2013, and references therein). Compared with older power plants, modern coal-fired power plants have substantially reduced SO_2 and primary particle emissions, yet their plumes seem to be very active in producing new aerosol particles that grow in size (Junkermann *et al* 2011b, Mylläri *et al* 2016). It is clear that more attention should be paid not only to sulfur, but also to other low-volatility compounds capable of contributing to NPF and subsequent particle growth in various point-source plumes.

4.2. Coastal zones and sea-ice regions

Coastal zones, i.e. the regions between the open oceans and continents or islands, display a very specific kind of local atmospheric NPF. The most thoroughly-studied coastal zone in this regard is Mace Head in the western coast of Ireland, where the most intense NPF was observed to take place in the simultaneous presence of a low tide and solar radiation (O'Dowd *et al* 1998, 1999). These NPF events were estimated to occur over the spatial scales of <100 m, and were believed to produce initially very rapidly-growing particles that reached sizes from a few nm up to 10–20 nm by the time they arrived at the measurement station (O'Dowd *et al* 1998, Dal Maso *et al* 2002, Ehn *et al* 2010). The coastal NPF around Mace Head was ascribed to photochemical reactions involving iodine compounds emitted by the algae exposed to air and sunlight in a narrow region near the coastline (O'Dowd *et al* 2002, Sipilä *et al* 2016).

In addition to Mace Head, intense coastal NPF have been observed in a few other locations: Bodega

Bay in California (Wen *et al* 2006), Cape Grim in Tasmania (Grose *et al* 2007), Great Barrier Reef on the east coast of Australia (Modini *et al* 2009), Roscoff in the northwest of France (Whitehead *et al* 2009, McFiggans *et al* 2010), and O Grove on the northwest coast of Spain (Mahajan *et al* 2011). In Cape Grim, Roscof and O Grove, coastal NPF was confirmed to be associated with iodine emissions, while low-tide conditions seemed to favor NPF in Roscoff and O Grove. In Bodega Bay, ocean upwelling bringing nutrients from subsurface waters seemed to be the most influential factor for NPF.

Another kind of environment next to a coastline with a high potential to produce new aerosol particles is sea ice, especially melting sea ice. Evidence for, or indications of, atmospheric NPF associated with air masses originating from partly sea-ice covered regions were found in measurements conducted over the Greenland Sea and in Svalbard, Arctic (Allan *et al* 2015, Dall'Osto *et al* 2017), as well as during several measurement campaigns made in and around Antarctica (Davison *et al* 1996, Atkinson *et al* 2012, Kyrö *et al* 2013, Roscoe *et al* 2015). In many of these studies, NPF was speculated to be initiated by iodine emissions from the melting sea ice.

4.3. Cloud-induced NPF

Indicative of NPF associated with clouds, several studies that were conducted already a couple of decades ago reported elevated total (Hegg *et al* 1990, Hegg *et al* 1991, Hudson and Frisbie 1991, Radke and Hobbs 1991) or sub-10 nm (Keil and Wendisch 2001, Weber *et al* 2001) particle number concentrations near cloud edges as compared with regions further away from cloud or inside the cloud. These observations were made in both marine and continental atmosphere, and for clouds ranging in altitude from the BL up to several km in the FT. The most common, but not exclusive, near-cloud regions for NPF seemed to be the air just above cloud tops. Several explanations for this phenomenon were suggested, including enhanced UV irradiance and turbulence in the vicinity of clouds (e.g. Wehner *et al* 2015).

Besides cloud edges, outflow regions of mainly convective clouds were found to be locations with active NPF (Perry and Hobbs 1994, Clarke *et al* 1998b, 1999, Ström *et al* 1999, Clement *et al* 2002, Twohy *et al* 2002, Waddicor *et al* 2012). Weigelt *et al* (2009) measured upper-troposphere particle number size distributions over different regions around the world, and found a frequent presence of nucleation mode particles whenever the measured air masses had been in recent contact with deep convective cloud systems. Similar observations had already been reported over the Indian and Pacific Oceans (de Reus *et al* 2001, Clarke and Kapustin 2002), and over the Amazon Basin (Andreae *et al* 2018). Theoretical analysis and model simulations support the view that clouds associated with deep convection provide favorable conditions for NPF, however the precursors, nucleation mechanisms and exact location of NPF with respect to clouds remain to be quantified (Kulmala *et al* 2006b, Ekman *et al* 2008, Waddicor *et al* 2012).

Finally, there is some evidence on NPF taking place in cirrus clouds (Lee *et al* 2004, Weigel *et al* 2011). Although the large CS inside any cloud is expected to strongly disfavor NPF, Kazil *et al* (2007) showed that conditions inside cirrus clouds allow the occurrence of sulfur-derived NPF.

4.4. NPF associated with transportation

Emissions associated with transportation are an important source of ultrafine aerosol particles into the atmosphere (Kumar *et al* 2013, Paasonen *et al* 2016). In emission inventories, all these particles are usually counted as primary aerosol particles, even though some fraction of them are small (<30 nm diameter) and volatile when heated up to 100 °C–300 °C, i.e. presumably formed by NPF in the atmosphere rapidly after the exhaust emissions are cooled and diluted into the ambient air. Below we discuss shortly the observed characteristics of NPF associated with three major transport sectors: ships, aircraft and motor vehicles.

Particle size distribution measurements in ship plumes show frequently the presence, and sometimes dominance, of volatile nucleation mode particles (e.g Petzold *et al* 2008, Lack *et al* 2009, Jonsson *et al* 2011, Pirjola *et al* 2014). The contribution of these particles to the total particle number concentrations seems to depend on the overall composition of the ship plume and its aging time in the atmosphere. Due to the potentially large adverse health effects caused by ship traffic near coastal zones, air quality regulations are gradually changing the properties of ship emissions (Corbett *et al* 2007, Fuglestvedt *et al* 2009, Liu *et al* 2016). While these changes will evidently reduce particle mass concentrations resulting from ship traffic, the corresponding influences on particle number concentrations are more complicated with even a possibility to increase the number of volatile (by heating) nucleation mode particles (Lack *et al* 2011, Petzold *et al* 2011).

Airplanes have been known to emit volatile nucleation mode particles into the lower troposphere and lower stratosphere for quite some time (Fahey *et al* 1995, Pueschel *et al* 1998, Schröder *et al* 1998, Anderson *et al* 1999). More recently, aviation was found to increase substantially total particle number concentrations in the vicinity of several major airports (e.g. Hudda *et al* 2016, Masiol *et al* 2017, Shirmohammadi *et al* 2017), and several studies reported clear evidence on NPF in the associated aircraft plumes (Herndon *et al* 2008, Mazaheri *et al* 2009, Kinsey *et al* 2010, Buonanno *et al* 2012, Timko *et al* 2013, Hudda and Fruin 2016). The relative importance of primary soot particle and volatile nucleation mode particles in the surface air affected by aviation operations remains, however, poorly quantified.

Motor vehicle traffic is an example of extremely localized NPF, mainly because the newly-formed particles are typically tied to the presence of individual vehicles. Particle number size distributions measured at traffic-impacted sites show frequently a dominant peak at particle diameters below 30 nm (see Vu *et al* 2015 and references therein), which indicates that motor vehicle traffic is a very strong source of nucleation mode particles in the urban atmosphere. Field and laboratory experiments have demonstrated that the presence and properties of nucleation mode particles in vehicle emissions depend on multiple factors, including the vehicle engine and emission after-treatment technologies, driving conditions, used fuels and lubricant oils, and ambient meteorological conditions (e.g. Lee *et al* 2015, Karjalainen *et al* 2016b, 2016c, Saliba *et al* 2017, Timonen *et al* 2017). A further complication arises from the observations that the nucleation mode particles associated with motor vehicle emissions may contain a non-volatile core (Sgro *et al* 2008, Lähde *et al* 2009). This means that some of these particles are formed inside the engine, not by NPF in the diluting exhaust plume. Finally, Rönkkö *et al* (2017) observed a frequent presence of a large number of sub-3 nm particles in locations exposed to road traffic. These nanocluster aerosol particles not only give an additional contribution to ultrafine particle populations, but they could also act as nucleating cores for particles causing a NPF and growth event in urban environments.

5. Growth of newly-formed particles to larger sizes

5.1. General features

Analyses of regional NPF events based on ion spectrometer measurements in different environments have revealed a clear size dependency in the GR of sub-20 nm diameter particles (Hirsikko *et al* 2005, Virkkula *et al* 2007, Suni *et al* 2008, Yli-Juuti *et al* 2009, Manninen *et al* 2010, Vakkari *et al* 2011, Yli-Juuti *et al* 2011, Hirsikko *et al* 2012, Gonser *et al* 2014, Dos Santos *et al* 2015, Rose *et al* 2015b, Kontkanen *et al* 2016a, Vana *et al* 2016). Most of these studies have considered three size ranges (1.5—3 nm, 3—7 nm and 7—20 nm) and observed that the average particle GR is the lowest in the sub-3 nm size range and the largest in the 7—20 nm size range. Exceptions for this pattern have been reported at two slightly elevated measurements sites (Hohenspessenberg in Germany and Pallas in Northern Finland; see Manninen *et al* 2010) and at a semi-clean site in Southern Africa (Vakkari *et al* 2011).

Apart from the almost universal summer maximum in average particle GRs (see section 3.2.2), there are some indications that also the size dependency of the GR might be at its strongest during the summer time (Yli-Juuti *et al* 2011, Vakkari *et al* 2011, Häkkinen *et al* 2013). These observations suggest that biogenic emissions play an important role in the growth of newly-formed particles, at least in the continental BL, and that most of the compounds contributing to the particle growth prefer larger particles over the small (<a few nm) ones. Such a size-selectivity could be explained by (1) different volatilities of the gas-phase oxidation products of biogenic vapors, so that only the least volatile ones are capable of condensing onto the smallest particles, or (2) by differences in the size dependency of the different particle growth processes, such as condensation growth and heterogeneous reactions either on particle surfaces or inside them (e.g. Pierce *et al* 2011, Riipinen *et al* 2012, Donahue *et al* 2013, Häkkinen *et al* 2013, Tröstl *et al* 2016, Apsokardu and Johnston 2018). These two explanations are not mutually exclusive, but might act simultaneously. Getting more insight into this issue would benefit from field data on whether and how the particle GR depends on the particle size above 20 nm diameter. Unfortunately, with the exception of a couple of recent studies indicating that Aitken and accumulation mode particles may growth faster than nucleation mode particles (Burkart *et al* 2017, Paasonen *et al* 2018), practically no data on this subject is available at the moment.

A few field studies have investigated in detail how the particle GR behaves in the sub-3–5 nm size range. While most of these studies report a strong increase in the particle GR with increasing particle size at this size range (Kuang *et al* 2012, Kulmala *et al* 2013, Xiao *et al* 2015), also more complicated patterns of this size dependency have been suggested (Yu *et al* 2016). More information on the GRs of the smallest newly-formed particles is needed for modeling purposes because these particles are most susceptible to coagulation scavenging (see section 2.1.2), and for understanding why NPF is possible even under extremely polluted conditions (Kulmala *et al* 2017).

5.2. Chemical compounds contributing to the particle growth

5.2.1. Direct particle composition measurements

Observations of aerosol particle chemical composition combined with simultaneous measurements of aerosol particle size distributions have been used to identify the chemical composition of the compounds responsible for particle growth in NPF events. Direct observations of nucleation-mode particle chemical composition have been made with thermal desorption chemical ionization mass spectrometer (TDCIMS) (Smith *et al* 2004) and with nano aerosol mass spectrometer (NAMS) (Wang *et al* 2006). Of these two instruments, NAMS, measures the atomic composition of the particles, while TDCIMS utilizes a softer ionization method and can identify the chemical composition of the particles in more detail (e.g. Smith *et al* 2010). However, as most studies do not report detailed composition of the condensable vapors, we will group the identified growth channels into growth by condensation of organic compounds (organic aerosol, OA), sulfate (SO_4), ammonium (NH_4) and nitrate (NO_3).

In addition to chemical composition measurements of the nucleation mode, also size-resolved chemical composition measurements for >30 nm with aerosol mass spectrometer (AMS) (Jayne *et al* 2000) and non-size resolved submicron aerosol chemical composition measurements with aerosol chemical speciation monitor (ACSM) (Ng *et al* 2011) have been utilized. In the AMS-based studies two approaches have been used to infer the chemical composition of the growth. If concentrations of the measured compounds are high enough to have good signal levels, the composition of <60 nm particles can be considered to represent the chemical composition once the newly-formed particles have grown above the AMS detection limit (e.g. Zhang *et al* 2004). On the other hand, the time evolution of the chemical composition of the bulk submicron aerosol can be used to infer the source rate for different chemical compounds if it can be assumed that changes in the submicron aerosol composition are dominated by condensation during the NPF event (e.g. Pierce *et al* 2011). In most cases, if the size time evolution of the number size distribution is smooth enough to derive formation and GRs during NPF event, also the source rates of OA, SO_4, NH_4 and NO_3 can be derived from the time derivative of the respective AMS mass concentrations (e.g. Vakkari *et al* 2015).

When comparing chemical composition measurements between the nucleation mode and larger sizes, the effect of volatility needs to be taken into account. This means that as particles grow into larger sizes, (organic) compounds with higher saturation vapor pressure are able condense onto them (Donahue *et al* 2011). A direct consequence of this is that estimating the growth of nucleation mode particles based on chemical composition measurements of Aitken and accumulation mode particles with an AMS causes an overestimation on the contribution of organic compounds to this growth (e.g. Ehn *et al* 2014). However, measuring Aitken and accumulation mode particles can be interpreted to represent the composition responsible for the particle growth to CCN-sizes.

Measurements of the composition of compounds contributing to growth have been carried out in diverse environments ranging from urban to remote continental areas. At urban locations, studies have been carried out at Pittsburgh, US (Zhang *et al* 2004), Beijing, China (Wiedensohler *et al* 2009, Zhang *et al* 2011), Bakersfield, US (Ahlm *et al* 2012), Wilmington, US (Bzdek *et al* 2012) and Brisbane, Australia (Crilley *et al* 2014). Additionally, Setyan *et al* (2014) reported observations made in the urban outflow of Sacramento, US, and Vakkari *et al* (2015) reported observations made in the urban outflow of Johannesburg, South Africa. All urban studies utilized AMS measurements combined with number size distribution measurements, except Bzdek *et al* (2012) where a combination of NAMS and number size distributions was used, and Vakkari *et al* (2015) where a combination of ACSM and number size distributions was used. An overview of the chemical composition of the growth at urban locations is given in figure 5. Two of the above-mentioned urban studies (Wiedensohler *et al* 2009, Crilley *et al* 2014) did not report quantitative composition, and are therefore not included in figure 5, nevertheless both these studies concluded that the growth was dominated by inorganic species.

Observations at regional background locations, representing typically a mixture of anthropogenic and biogenic sources, include Tecamac, Mexico (Smith *et al* 2008), Lewes, US (Bzdek *et al* 2012, 2013, 2014) and Welgegund, South Africa (Vakkari *et al* 2015). The study at Tecamac is based on TDCIMS measurements, the studies at Lewes on NAMS measurements and the study at Welgegund on ACSM measurements. Similar to urban locations, the two main constituents of particle growth at regional sites are OA and SO_4 (figure 5). Likewise, a study utilizing AMS measurements at Melpitz, Germany (Wu *et al* 2015) identified SO_4, NH_4 and OA as the main compounds responsible for the particle growth, but the fractional contribution of each compound was not estimated. A recent study with TDCIMS at Southern Great Plains, US (Hodshire *et al* 2016) determined the contributions of OA, SO_4, NH_4 and amines to the particle growth.

In contrast with urban and regional studies, measurements in the environments with least anthropogenic influence indicate the growth dominated by OA ('rural' in figure 4). In this category, we consider observations from Hyytiälä, Finland (Allan *et al* 2006, Pierce *et al* 2011, Riipinen *et al* 2012, Pennington *et al* 2013), Egbert, Canada (Pierce *et al* 2011), Whistler Mountain, Canada (Pierce *et al* 2012) as well as the clean sectors at Cool, US (Setyan *et al* 2014) and at Welgegund, South Africa (Vakkari *et al* 2015). Also Han *et al* (2014) report OA-dominated growth from Wakayama, Japan. Except for Pennington *et al* (2013) all studies from the rural locations are based on AMS or ACSM measurements.

In summary, the chemical composition of the growing particles depends on the mixture of precursor compounds present at the time of the NPF event. Consequently, as more and more pristine environments are considered, the fraction of OA increases. This is seen clearly at sites that have distinct clean sectors: at a single location the growth can vary from fully OA to SO_4-dominated depending on the air mass origin (Setyan *et al* 2014, Vakkari *et al* 2015, Hodshire *et al* 2016). Finally, it is worth noting that amines have been observed in nucleation and Aitken mode particles at several locations, even though their fractional contribution to the particle growth has not been

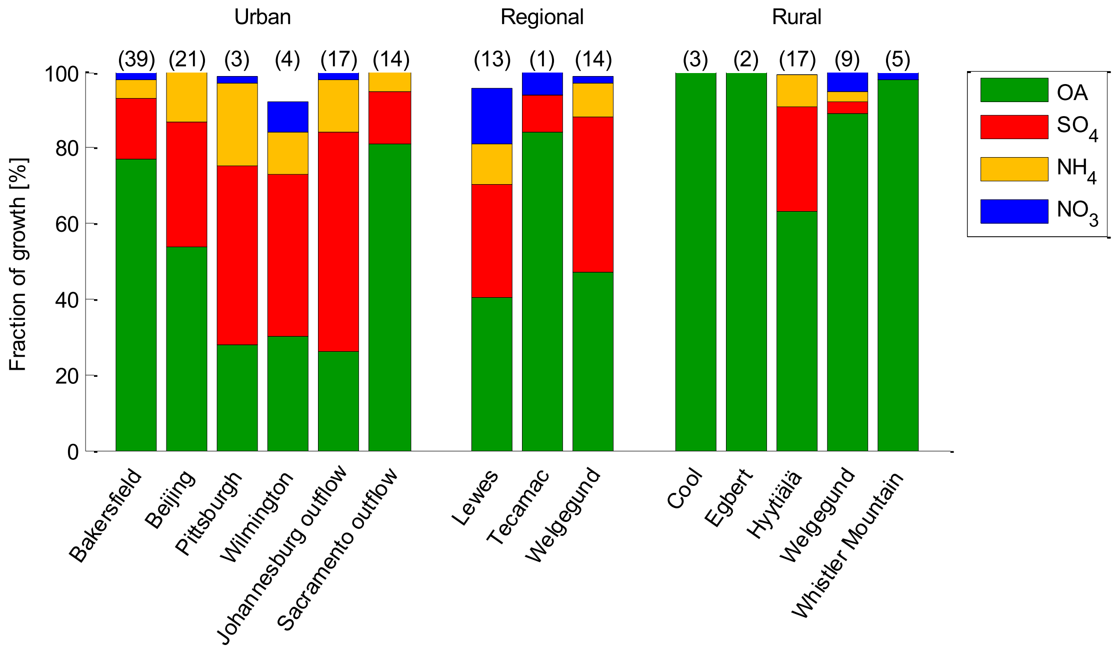

Figure 5. Mean chemical composition of the growth reported for urban, regional background and rural locations (see the text in section 5.2.1 for a complete set of references). The number of analyzed NPF events is indicated for each location. For Lewes and Hyytiälä, several papers report direct chemical composition measurements of particle growth; these records have been averaged in this graph. Note that some studies (Pierce *et al* 2011, Zhang *et al* 2011, Setyan *et al* 2014) report only the source rates of OA and SO_4, that on average 8% of the elemental composition of the nucleation mode was left unapportioned at Wilmington (Bzdek *et al* 2012), and that on average 3% of nucleation mode composition was apportioned to SiO_2 at Lewes with a smaller fraction left unapportioned (Bzdek *et al* 2014). For this comparison, SO_4 in these studies is assumed to be fully neutralized by NH_4.

quantified (Mäkelä *et al* 2001, Smith *et al* 2010, Hodshire *et al* 2016).

5.2.2. Indirect observations

The two most widely used indirect approaches to get information on the compounds responsible for the particle growth during atmospheric NPF are to measure the particle hygroscopicity or volatility, or some combination of these two. Hygroscopicity measurements reveal the particle mixing state with respect to their water uptake and are useful in distinguishing between totally water-insoluble material, highly-hygroscopic substances like sulfuric acid or ammonium sulfate, and less-hygroscopic material like a big fraction of secondary organic compounds. Volatility measurements provide additional insight into this chemistry, as compounds with different vapor pressures are expected to be evaporated from particles at different temperatures. Both hygroscopicity and volatility measurements can be conducted with a good time resolution and for several pre-selected particle sizes (e.g. Swietlicki *et al* 2008, Villani *et al* 2008).

Hygroscopicity and volatility measurements made in forested environments indicate that both highly- and less-hygroscopic material condense onto particles formed by atmospheric NPF, and that the contribution of highly-hygroscopic material is more important for smaller particles and during the photochemically active time of the day (Hämeri *et al* 2001, Petäjä *et al* 2005, Ehn *et al* 2007a, Ristovski *et al* 2010, Wu *et al* 2013, Jung and Kawamura 2014). In urban environments, the relative contributions of highly- and less-hygroscopic material to the particle growth seem to vary, in addition to which nucleation mode particles show occasionally an external mixture indicative of two very different sources for these particles (Väkevä *et al* 2002b, Sakurai *et al* 2005, Petäjä *et al* 2007, Park *et al* 2009). The few measurements made in coastal environments show a relatively high contribution of highly-hygroscopic material in nucleation mode particles and indications of almost insoluble material in the smallest particles (Väkevä *et al* 2002a, Johnson *et al* 2005, Park *et al* 2009). Finally, an investigation conducted at a high-latitude Arctic site, Svalbard, suggests a very large fraction of ammonium sulfate in particles originating from atmospheric NPF (Giamarelou *et al* 2016).

In a couple of locations, particle volatility measurements have revealed the presence of a practically non-volatile core in growing nanoparticles (Wehner *et al* 2005, Ehn *et al* 2007b, Wang *et al* 2017b). This core was found to occupy about 20%–40% of the particle volume, with no apparent size dependency for this fraction. The origin of the non-volatile core is somewhat unclear but was hypothesized to result from either photochemical or heterogeneous processes that produce extremely non-volatile material into the growing particles (Wang *et al* 2017b).

In addition to particle hygroscopicity and volatility measurements, a few other experimental techniques have been developed and applied over the years to get information about the chemical composition of nucleation mode particles in the atmosphere. Such techniques include the transition electron microscopy which provides morphological information of particles (e.g. Mäkelä *et al* 2002, Leck and Bigg 2005, Karl *et al* 2013), pulse height analyzer ultrafine condensation particle counter used in parallel with mobility size distribution measurements (O'Dowd *et al* 2003), ultrafine organic tandem differential mobility analyzer which provides an estimate on the organic fraction of particles of pre-selected sizes (Vaattovaara *et al* 2005, Laaksonen *et al* 2008), and condensation particle counter battery which reveals the water-affinity of sub-20 nm particles (Kulmala *et al* 2007, Riipinen *et al* 2009). While providing some new insight into this topic, the information obtained using these approaches has, in most cases, remained rather qualitative.

A few studies have measured gas-phase concentrations of one or more LVOC compounds and, by assuming their irreversible condensation onto aerosol particles, then estimated how big fraction of the observed particle growth can be explained by these compounds. In case of gaseous sulfuric acid, this fraction was found to be below 10%, on average, at two forested European sites with very different levels of exposure to anthropogenic pollutants (Boy *et al* 2005, Fiedler *et al* 2005). A much higher contribution by sulfuric acid, between about 30% and 60%, was reported for the particle growth in Beijing, China (Yue *et al* 2010). Since the pioneering studies by Ehn *et al* (2012, 2014), people have gradually started to measure gas-phase concentrations of a subset of highly oxygenated (organic) molecules (HOMs), or their clusters, under field conditions (Mutzel *et al* 2015, Jokinen *et al* 2016, Bianchi *et al* 2017, Frege *et al* 2017, Mohr *et al* 2017). HOMs include both ELVOCs and LVOCs (Tröstl *et al* 2016), so they are very good candidates for the organic compounds that give a major contribution to the growth of newly-formed particles in the atmosphere. Quantitative estimates on how big fraction of the observed particle growth can be explained by different HOMs, or groups of HOMs, do not yet exist for real atmospheric aerosol systems. Finally, it should be noted that a number of studies have estimated the gas-phase concentration of sulfuric acid or (extremely) LVOC organic compounds from direct measurements of their precursor compounds in the gas phase using either available proxies for this purpose (e.g. Petäjä *et al* 2009, Mikkonen *et al* 2011, Kontkanen *et al* 2016c) or by simulating explicitly the precursor gas-phase chemistry, and then used these values to further estimate the particle GR. Most of these studies agree in a broad sense with the results presented above but, because of the inherently large uncertainties associated with these analyses, will not be reviewed here.

5.3. Implications for the particle number and CCN budgets

In addition to the data obtained using various modeling frameworks mentioned in section 1, the contribution of atmospheric NPF to the total or ultrafine particle number concentration, or to the CCN budget, can be estimated from atmospheric measurement data. Below we summarize shortly the main results obtained from such measurements.

Rodriguez and Cuevas (2007) introduced a method by which one can estimate the contribution of NPF to the total particle number concentration based on simultaneous aerosol black carbon and particle number concentration or size distribution measurements. This method was applied to several urban areas in Europe, and the average contribution by NPF was found to vary between about 20% and 70% with most values being slightly above 50% (Rodriguez and Cuevas 2007, Fernandez-Camacho *et al* 2010, Reche *et al* 2011, Hama *et al* 2017). Kulmala *et al* (2016b) refined the approach proposed by Rodriguez and Cuevas (2007) and investigate separately the different particle size modes and associated uncertainties. They estimated that on average, NPF contributes about 70% and 80% to the total particle number concentration in a heavily-polluted urban site and rural forested site, respectively. At both sites, these contributions were estimated to be the lowest in the accumulation mode, somewhat higher in the Aitken mode and clearly the highest in the nucleation mode. All these numbers should be interpreted with some caution due to the inherent uncertainties in the applied method. For example, the method classifies a large fraction of such primary particles that contain little or no BC as particles formed by NPF, which can be questionable for small combustion sources like emissions from individual vehicles (see section 4.4).

By using various kinds of source apportionment methods, the average contribution of photochemical NPF to the total particle number concentration was estimated to be 3% at an urban background site in Barcelona, Spain (Pey *et al* 2009), 8% in urban St. Louis, Missouri, US (de Foy and Schauer 2015), 4% and 25% in Ausburg, Germany, and Rochester, New York, US, respectively (Vu *et al* 2015), 17% in central Los Angeles, US (Sowlat *et al* 2016), and 11% (2001–2002) and 6% (2016–2017) in Pittsburg, Pennsylvania, US (Saha *et al* 2018). These values are lower than those obtained using the approach presented in the previous paragraph, and at this stage it is impossible to judge whether this feature is real or a sign of problems in one of the used approaches. Finally, three studies estimated the contribution of NPF to the ultrafine particle number concentration by applying sophisticated methods to a diurnal evolution of the particle number size distribution: Ma and Birmili (2015) reported this contribution to be 7%, 14% and 30% for roadside, urban background location and regional background

Table 2. Studies estimating the strength at which atmospheric NPF produce CCN based on long-term observations. Three different methods have been used to determine CCN concentrations: CCN counter measurements (CCNC), particle number size distribution measurements (SD) and combined particle number size distribution and hygroscopicity measurements (SD + H). The probability gives the fraction of NPF events that were estimated to produce CCN in these studies. EF is the reported average enhancements factors in CCN concentrations due to individual NPF and growth events. The range of values in EF reflect different water vapor supersaturations for CCN (CCNC) or effective cut-off diameters above which all larger particles have been assumed to be CCN (SD and SD + H).

References	Site(s)	Method	Probability	EF
Lihavainen *et al* (2003)	Remote	SD	18%	4.1–11
Kuang *et al* (2009)	2 urban + rural	SD	—	3.8
Asmi *et al* (2011)	Remote	SD	—	1.5–3.8
Sihto *et al* (2011)	Rural	CCNC	—	0.7–1.1
Yue *et al* (2011)	Urban	SD + H	—	0.4–6
Laakso *et al* (2013)	Rural	SD	—	>0.5
Wang *et al* (2013)	Urban	SD	—	5.5–7.8
Yu *et al* (2014)	Rural	SD	—	4.7
Shen *et al* (2016b)	Rural	SD	17%	3.9–4.7
	Mountain	SD	12%	3.0–3.8
	Regional background	SD	15%	2.6–3.7
Dameto de Espana *et al* (2017)	Urban	CCNC	~40%	<1.4
Rose *et al* (2017)	Mountain	SD	61%	—

site, respectively in Leipzig, Germany, while Salma *et al* (2017) reported it to be 13% and 37% in the city center and near-city background site, respectively, in Budabest, Hungary. Nemeth *et al* (2018) found that compared with central Budabest, the contribution of NPF to the ultrafine particle number concentrations is probably slightly lower at an urban background site in Vienna, Austria, but clearly larger at an urban background site in Prague, Czech Republic.

From the climate point of view, the importance of atmospheric NPF manifests itself in the growth of newly-formed particles to the sizes at which they can act as CCN. Kerminen *et al* (2012) reviewed the available literature on this subject and concluded that, depending on the cloud maximum water vapor saturation ratio and chemical composition of the growing particles, they need to reach sizes between about 50 and 150 nm in diameter to participate in cloud droplet activation. Particles smaller than 50 nm can only be activated at exceptionally high water vapor saturation ratios, such as those encountered in deep convective cloud systems (Fan *et al* 2018). The growth of newly-formed particles up to a few tens of nm can be completed within a few hours, but commonly such growth requires more than a day of atmospheric ageing. Over such time scales, it is observationally very challenging to separate between CCN originating from atmospheric NPF and CCN originating from the growth of small primary aerosol particles (Kerminen *et al* 2012).

A number of papers have analyzed case studies on observed CCN production associated with atmospheric NPF. Such studies cover forested areas in Canada (Leaitch *et al* 1999, Pierce *et al* 2012, Shantz *et al* 2012) and Japan (Han *et al* 2013), other rural areas in central Europe (Wu *et al* 2015) and United States (Creamean *et al* 2011, Levin *et al* 2012), high-altitude mountain site (Friedman *et al* 2013), Arctic Archipelago and Ocean (Willis *et al* 2016, Burkart *et al* 2017), Mediterranean environment (Kalivitis *et al* 2015) and sites experiencing different degrees of anthropogenic influence in China (Wiedensohler *et al* 2009, Leng *et al* 2014, Li *et al* 2015a, Ma *et al* 2016, Yue *et al* 2016, Li *et al* 2017). These studies, while focusing on varying aspects of this phenomenon, illustrate that atmospheric NPF can lead to CCN formation in vastly different atmospheric environments.

Using long-term observations, several studies have attempted to estimate how frequently atmospheric NPF leads to CCN production and/or how much NPF events enhance CCN concentrations. Table 2 summarizes these studies and shows that, depending on the location, from about 10% to 60 % of the observed NPF events led to the production of new CCN. Furthermore, compared with the situation before a NPF event, CCN concentrations were found to be enhanced by up to several hundred per cent after the newly-formed particles had grown to larger sizes. Peng *et al* (2014) combined observations from several sites in China and found that the fraction of NPF events leading to CCN production was clearly the highest in summer and close to zero in winter. The increase in CCN concentrations caused by NPF seem to have a complicated seasonal pattern, as demonstrated by Kerminen *et al* (2012) for three rural/remote sites in Europe and a rural site in South Africa. A couple of studies estimated the contribution of NPF to the overall CCN budget, being about 30% for a polluted rural site in Europe (Laaksonen *et al* 2005) and about 50% for a forested site in Canada (Pierce *et al* 2014).

Very few studies have compared observed atmospheric CCN production resulting from NPF to that obtained from model simulations (Laakso *et al* 2013, Westervelt *et al* 2013, Cui *et al* 2014). While these studies demonstrated some success in simulating the growth of newly-formed particle into CCN, they also brought up several deficiencies in our current understanding on this phenomenon. Noting this and the observational challenges in estimating the role of atmospheric NPF in the CCN budget, as discussed above, we conclude that it would be essential to perform studies that combine model simulations with information obtained from both field measurements and laboratory experiments.

6. Summary and outlook

6.1. Main findings

While the spatial and temporal coverage of atmospheric NPF evidently varies over several orders of magnitude, it has become customary to divide this phenomenon into two broad categories: regional and sub-regional NPF. Regional NPF takes simultaneously place over distances of several tens to hundreds of kilometers, even though with a variable intensity in both time and space. The large spatial coverage of regional NPF makes it often possible to determine several quantities characterizing the timing and intensity of this phenomenon by using continuous measurements made at a fixed location within the zone of NPF. Ideally, these quantities include the starting time and duration of NPF that together define a so-called NPF event, the frequency of occurrence of NPF events, as well as the formation and GRs of particles during a NPF event. The spatial extent of sub-regional NPF varies from a few meters in vehicle plumes to about a few kilometers, or even tens of kilometers, in plumes from major point sources, such as power plants and oil refineries. For sub-regional NPF, atmospheric measurements can usually provide only rather limited information about the timing and intensity of this phenomenon.

Atmospheric NPF is very strongly a daytime phenomenon, as observations of nighttime NPF have been reported from less than 10 measurement sites so far. The typical duration of a regional NPF event is a few hours, and usually no more than one NPF event per day is being observed. The frequency of occurrence of NPF varies over the course of the year, with the tendency of its maximum to shift from summer in polar and many high-latitude regions toward spring or autumn in most other regions. Only few long-term NPF observations exist, and the existing data seems to indicate a substantial inter-annual variability in the NPF frequency. A notable decline in the frequency of occurrence of NPF events over the years was reported for a few sites that had experienced substantial reductions in the ambient SO_2 concentration.

Regional NPF seems to take place across all continental environments in the lower troposphere, even though with a highly varying frequency. The observed factors that favor the occurrence of regional NPF include a high intensity of solar radiation, low RH, high gas-phase sulfuric acid concentration, and low pre-existing particle loading, i.e. low CS and CoagS. The ambient temperature and SO_2 concentration appear to be important quantities as well, but their roles remain ambiguous because they influence both the factors that enhance NPF and the factors that suppress it. Sub-regional NPF has been observed to take place in anthropogenic plumes originating from various ground-based point sources, in biomass burning plumes, near cloud edges and in cloud outflow regions, as well as in some coastal areas and ice-melting regions. Small-scale NPF is also common in vehicle, ship and aircraft plumes, even though in emission inventories aerosol particles originating from these sources are commonly considered as primary particles. The occurrence and intensity of sub-regional NPF depends, in addition to the same factors that influence regional NPF, also on atmospheric dispersion conditions and on fuels or burning materials as well as combustion conditions in the case of combustion sources.

Up to the present, particle formation rates (J) and GRs associated with regional NPF have been reported for more than 200 measured sites. We determined the statistics of these quantities separately for the following site categories: boreal forests, Arctic regions, Antarctica, mountain sites, other remote and rural areas, urban environments, as well as rural, suburban, urban and marine/coastal areas in China. We found that, within these site categories, the median value of J is about 0.05 $cm^{-3}\,s^{-1}$ in Antarctica, slightly below 1 $cm^{-3}\,s^{-1}$ in both Arctic and boreal regions, between about 3 and 5 $cm^{-3}\,s^{-1}$ in other rural and remote environments as well as in urban areas outside China, and about 8 $cm^{-3}\,s^{-1}$ in urban China. The median value of GR has a much smaller variability between the different site categories, being in the range of 2.3–7.4 $nm\,h^{-1}$.

The rate at which newly-formed particles grow to larger sizes has been observed to increase with an increasing particle size in the sub-20 nm particle size range, especially during summer, and there are some indications that this pattern may continue for larger particle sizes. At most of the measurement sites, the growth of particles to larger sizes, along with the size dependency of this GR, appears to be dominated by the uptake of organic vapors by these particles. In rural environments sulfuric acid and associated bases like ammonia have been estimated to explain a few per cent or less of the observed particle growth. On the other hand, in urban environments sulfuric acid and ammonia may explain up to 70% of the observed particle growth.

An increasing number of observational studies have been dedicated to estimate the contribution of atmospheric NPF to number concentrations of total aerosol particles, ultrafine particles or CCN. The vast majority of the studies focusing on the ultrafine particle number budget have been conducted in urban areas, where from a few per cent up to about 70% of ultrafine particles were estimated to originate from NPF. It remains, however, still unclear to which extend this value range can reflect a true variability of particle sources between different urban environments, and to which extend it is affected by the application of different approaches to estimate the contribution of NPF. Observational studies on CCN production associated with atmospheric NPF cover a broad range of environments. These studies indicate that typically between about 10% and 60% of the observed NPF events can lead to a production of new CCN and, once this occurs, the CCN number concentration may increase by up to several folds. Altogether, atmospheric observations support the current view, obtained from large-scale model simulations, that atmospheric NPF is an important source of CCN in the global troposphere.

6.2. Future outlook

The interest in atmospheric NPF lies in the potential ability of this phenomenon to increase ultrafine particle number and CCN concentrations in spatial scales ranging from urban air to the global atmosphere. In order to quantify the contribution of NPF to ultrafine particle number or CCN budgets, one has to (i) understand which factors determine the occurrence of NPF, as well as its spatial and temporal extent, in different atmospheric environments, and (ii) have predictive models by which one can estimate the strength of NPF, more specifically the NPF and GR, in these environments. Atmospheric observations, when combined with suitable theoretical frameworks and information obtained from laboratory experiments, are crucial components in working toward improved understanding and predictive models. Below we summarize shortly our view on future needs for such observations and their analyses.

The apparent ubiquity and heterogeneity of atmospheric NPF requires long-term continuous observations at fixed locations that would ideally cover all the continents and major ecosystem types, different urban centers and mountain regions, as well as remote islands surrounded by different oceanic areas. In this regard, we are still lacking suitable observational data from large continental areas in Africa, Southern America, Asia and Australia, as well as from the vast majority of oceanic areas (figure 4). Without going into any details in the required instrumental techniques, continuous observations should include particle number size distribution measurements down to at least a few nm, and preferably down to 1–2 nm, in particle diameter as well as measurements of basic meteorological variables, including solar radiation intensity. To the extent possible, it would be useful to measure concentrations of the vapors that potentially participate in NPF and subsequent particle growth (H_2SO_4, ELVOCs, LVOC, ammonia and amines), concentrations of the precursor compounds for these vapors (SO_2, VOCs) as well as relevant oxidants. It should be kept in mind that in order to take the best possible advantage of continuous observations of NPF-related variables in further analyses, these observations should cover at least one full annual cycle in time. However, also longer time series of such observations are needed, since observed trends in the characteristics of atmospheric NPF are currently available from very few sites only (see section 3.1.2).

Urban areas have emerged as environments where atmospheric NPF needs to be considered because of its potentially important contribution to the ultrafine aerosol population and related health effects of ambient aerosol particles. As discussed in section 3.2.2, the frequency, intensity and seasonal timing of NPF vary a lot between different urban environments. This feature is understandable when considering that different urban areas probably have very distinct mixtures of vapors from each other that potentially participate in NPF and subsequent particle growth, as well as different combinations of the factors that tend to suppress NPF (e.g. high pre-existing particle loading). Another, yet poorly understood feature in this regard is the occurrence of regional NPF under highly-polluted conditions, which should not be possible based on our current theoretical understanding on atmospheric NPF (Kulmala *et al* 2017, see also section 3.1.1). The challenge for future work arising from these features is that, without more observational data from different urban areas and improved theoretical understanding, it may be difficult to generalize NPF-related findings from one urban area to another. The heterogeneous nature of sources for aerosol particles and their precursor compounds within any individual urban area, as well as the existence of many other sources producing ultrafine particles into urban air, lead to additional research questions that need to be answered in future work: how do we separate particles associated with NPF in urban air from ultrafine particles coming from other sources?, how should we deal with the fact that both local and regional NPF can be simultaneously taking place in urban air?, and how all this should be taken into account when categorizing and analyzing NPF events in urban environments?

The upper FT appears to be an active source of new aerosol particles into both FT and BL (sections 3.2.4 and 3.2.5). There are several research questions concerning NPF in the upper FT that have not been fully answered yet: how frequent and widespread is this phenomenon outside tropical areas?, what is the role of clouds in free-tropospheric NPF?, does it require ingredients from surface emissions to take place?, and if it does, how sensitive is it to the chemical characteristics of surface emissions and to the way these emissions are transported to the upper FT? Even less is known about NPF taking place at the interface between the FT and BL. Although this phenomenon has been observed to be common at several mountain sites, whether and to which extent it occurs outside the mountainous areas, as well as its overall contribution to regional and global aerosol populations, are yet to be explored.

The methods used for analyzing atmospheric NPF require further developments. First, the large fraction of measurement days categorized as 'undefined' in many of the published data sets on regional NPF indicates that we may need to rethink how to classify bursts of atmospheric NPF. This concerns all the environments, but is particularly important for NPF taking place in urban areas. Second, instead of determining event-average particle formation and GRs, we clearly need means to determine these quantities at a maximum time resolution of a few minutes during NPF and the subsequent particle growth. Third, since atmospheric NPF is evidently an important source of CCN in different environments, we need methods capable of determining size-dependent particle GRs from sub-3 nm sizes up to 100–200 nm in particle diameter. Finally, although this review does not discuss the initial steps of NPF, it is clear that understanding processes that take place in the sub-3 nm size range is very important and requires further refinements of methods used to characterize these processes.

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