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Key Points:

- Atmospheric ice-nucleating particles (INPs) play a critical role in weather and climate by facilitating ice formation in clouds
- This review summarizes current knowledge on observational constraints, modeling, and cloud impacts of INPs
- Research priorities are identified to both advance fundamental understanding and bridge the observation-model gap for INPs

Supporting Information:

Supporting Information may be found in the online version of this article.

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Ice-Nucleating Particles That Impact Clouds and Climate: Observational and Modeling Research Needs

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Abstract Atmospheric ice-nucleating particles (INPs) play a critical role in cloud freezing processes, with important implications for precipitation formation and cloud radiative properties, and thus for weather and climate. Additionally, INP emissions respond to changes in the Earth System and climate, for example, desertification, agricultural practices, and fires, and therefore may introduce climate feedbacks that are still poorly understood. As knowledge of the nature and origins of INPs has advanced, regional and global weather, climate, and Earth system models have increasingly begun to link cloud ice processes to model-simulated aerosol abundance and types. While these recent advances are exciting, coupling cloud processes to simulated aerosol also makes cloud physics simulations increasingly susceptible to uncertainties in simulation of INPs, which are still poorly constrained by observations. Advancing the predictability of INP abundance with reasonable spatiotemporal resolution will require an increased focus on research that bridges the measurement and modeling communities. This review summarizes the current state of knowledge and identifies critical knowledge gaps from both observational and modeling perspectives. In particular, we emphasize needs in two key areas: (a) observational closure between aerosol and INP quantities and (b) skillful simulation of INPs within existing weather and climate models. We discuss the state of knowledge on various INP particle types and briefly discuss the challenges faced in understanding the cloud impacts of INPs with present-day models. Finally, we identify priority research directions for both observations and models to improve understanding of INPs and their interactions with the Earth System.

Plain Language Summary Atmospheric ice-nucleating particles (INPs) are rare particles that play a critical role in enabling ice crystals to form in clouds. Ice crystals in clouds act as seeds for most precipitation that reaches the Earth's surface, and impact climate by changing the amount of sunlight clouds reflect. A variety of naturally occurring particle sources contribute to atmospheric INPs, including wind-blown dusts, sea spray particles, biological particles such as fungal spores, bacteria and pollen, and ash and other particles from forest fires. Human-caused particulate pollution may also contribute to atmospheric INPs in some circumstances. A better understanding of these particles, including their sources to the atmosphere, their interactions with atmospheric processes, and their impacts on clouds, is therefore required to improve climate predictability in the coming century. While impressive progress has been achieved in recent years in process-level understanding of INPs and their cloud impacts, progress in this area will require better integration between observational and modeling tools and perspectives. This review discusses the current state of knowledge regarding INPs, key observational and modeling gaps, and identifies priority research areas that emphasize a unified effort between measurement and modeling communities.

1. Introduction and Motivation

Atmospheric ice-nucleating particles (INPs) are required to initiate ice formation in clouds with temperatures between −38 and 0°C, and can influence the formation of ice crystals in clouds below −38°C. Through their impact on freezing and deposition, INPs influence the rate at which the initial—or primary—ice forms in clouds.

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Primary ice formation, in turn, initiates subsequent cloud microphysical and dynamic processes—including secondary ice production—that ultimately influence cloud structure, extent, radiative properties, weather, and climate. INPs are extremely rare in the atmosphere, with concentrations that can vary over several orders of magnitude, for example, from fewer than 0.01 to more than 100 L^{-1} for INPs active at −30°C (DeMott et al., 2010). Both the rarity of INPs and their high sensitivity to aerosol chemical and physical properties create difficulties in their measurement and characterization. Recent decades have seen a resurgence in research on atmospheric INPs (DeMott et al., 2011; Kanji et al., 2017), which has improved understanding of INP measurement methods and broadened the scope of observations (Cantrell & Heymsfield, 2005). Field experiments have demonstrated that introducing INPs into supercooled clouds can change the timing and location of precipitation (French et al., 2018). Models have shown significant impacts of INPs on clouds that can explain some observed variations in cloud properties (Bangert et al., 2012; Prenni, Harrington, et al., 2007; Vergara-Temprado et al., 2018). Top-down evidence from satellites and lidars suggests INPs are an important contributing factor in determining cloud phase (Choi et al., 2010; Zhang et al., 2012; Zhao et al., 2019). Substantial uncertainties remain, but these advances in experiment, observation, and theory have built confidence that the abundance of INPs can affect mixed-phase cloud properties, and their interactions with planetary radiation and precipitation.

A long-term grand challenge for the scientific community in this area is achieving the capability to confidently represent INPs and their impacts on clouds and precipitation in the numerical regional and global cloud models used to study weather and climate. This capability would unlock new insights into the feedbacks between aerosols, clouds, and climate, as well as into potential approaches to weather modification. However, many challenges remain, including the fundamentally multiscale nature of the problem, and the improvements required to both observational capabilities and modeling of physical processes. Recent review articles have discussed challenges associated with methods for measurement of INPs, the availability of INP measurements from different environments, ice nucleation terminology, and the representation of INPs in cloud microphysical models (Coluzza et al., 2017; Hiranuma et al., 2015; Hoose & Möhler, 2012; Kanji et al., 2017; Murray et al., 2012; Vali et al., 2015).

Another critical obstacle toward achieving this vision, which thus far has received less focused attention, is the significant remaining gap between current measurement-based insights into the particle sources of INPs from field and laboratory measurements, and the representations of INPs in large-scale models. This observation-model gap and the research advances that will be required to close it are the focus of this review. To advance research in this area, the observational community needs guidance on which types of measurements will offer the most value in providing inputs and constraints for models. Meanwhile, the modeling community needs clarification on which INP parameterizations are physically well-understood, and how to most appropriately apply INP parameterizations within large-scale atmospheric models. Studies that collaboratively aim to better constrain and characterize important aerosol and cloud processes are needed in order to advance our scientific understanding of aerosol-cloud interactions.

To provide context and a broad overview, Figure 1 summarizes some of the key challenges in both modeling and measurements of aerosol particles, INPs, and cloud processes. This paper focuses primarily on the challenges in bridging the gap between observations and modeling of aerosol particles and INPs (the lower two sections of Figure 1). Specifically, the scientific community will have bridged the observation-model gap for INPs and their aerosol sources when we can demonstrate two key achievements across a variety of meteorological conditions and environments:

1. **Closure between observed aerosol and observed INPs**: Given appropriate measurements of the aerosol size distribution and composition, we can achieve a good agreement between the predicted temperature- and humidity-dependent concentration of INPs and those observed by a separate measurement, across the full range of atmospherically relevant temperature and humidity conditions.
2. **Predictive understanding of INPs for atmospheric modeling**: Atmospheric models represent the key processes driving spatial and temporal variability in INP concentrations and are capable of hindcasting (when using constrained meteorology) and/or forecasting INP number "skillfully," that is, with objectively greater accuracy than a "naïve" model such as a climatological value.

While significant challenges also remain in the area of cloud models and observations, they are not the primary focus of this review. We refer readers to the other reviews cited herein for deeper discussions of specific issues, including cloud processes, INP measurements, and molecular-level processes impacting INPs.

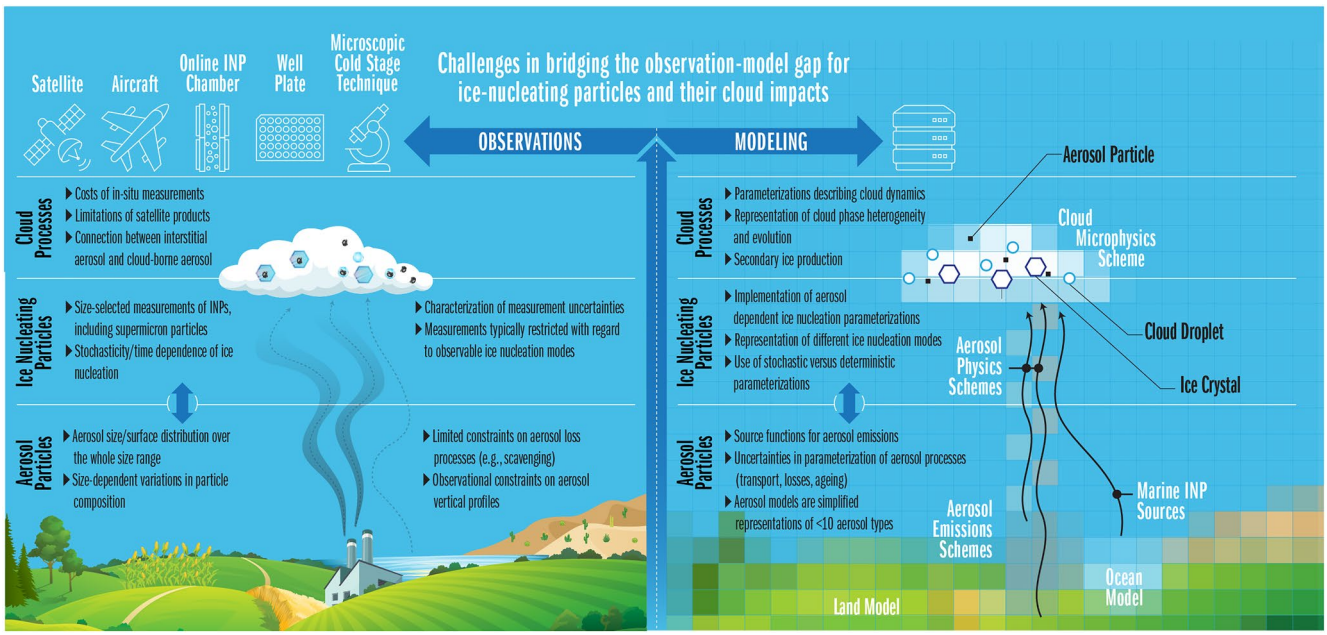

Figure 1. Overview of challenges in bridging the observation-model gap for aerosols, INPs, and their cloud impacts. On the left, observational challenges are listed over a cartoon environment. On the right, modeling challenges are listed over a cartoon of a visualization of a model "world" that approximates the earth system in grid boxes such that clouds are more box-like and cloud ice crystals are spherical.

After a brief overview of the current state of knowledge regarding the parameterization of INPs (Section 2), we discuss challenges in achieving both these aims. We begin with observational aerosol-INP closure in laboratory and field measurements (Section 2), then discuss predictive understanding of INPs (Section 3) as it relates to aerosol and cloud modeling at regional and global scales. Finally, we summarize key gaps and research needs for the community (Section 4).

2. Ice Nucleation Parameterizations

Ice nucleation parameterizations, developed on the basis of laboratory or field measurements, provide a key link between observations of INPs and their representation in atmospheric models. Due to constraints on both computing resources and physical understanding, for many decades ice nucleation in atmospheric models was largely predicted based on thermodynamic state variables such as ice supersaturation and temperature (Bigg, 1953; Meyers et al., 1992; Young, 1974). During the past decade, improved understanding of the dependence of ice nucleation on aerosol, as well as improved capabilities to simulate aerosol within atmospheric models, have increasingly led model developers to incorporate aerosol-aware parameterizations of ice nucleation (see Section 3.1).

Broadly, parameterizations of ice nucleation fall into two categories: "deterministic" parameterizations, where freezing behavior is modeled as a function of temperature, humidity, and aerosol properties (e.g., size, surface area, and chemical composition), and "stochastic" parameterizations, which additionally include a functional dependence on time (Vali et al., 2015). In the recent literature, deterministic parameterizations typically signify the effects of particle surface physicochemical properties on freezing and are usually represented by the "surface active site density" parameter ($n_s(T, q)$), which is a function of the temperature, T, and humidity, q (Ullrich et al., 2017). Meanwhile, time-dependent parameterizations typically are variations on classical nucleation theory (CNT; Knopf et al., 2020; Murray et al., 2012) and represent particle surface effects through the nucleation rate coefficient $J_{het}(T, q)$. Particle-type dependent parameterizations of either $n_s(T, q)$ or $J_{het}(T, q)$ are obtained for one or more of the modes of nucleation (see Box 1) through empirical fits to laboratory or field data (DeMott et al., 2015; Kanji et al., 2017; Knopf & Alpert, 2013; McCluskey, Ovadnevaite, et al., 2018; Murray et al., 2012). Important practical challenges associated with using INP parameterizations in atmospheric models will be discussed in Section 3.1.

Box 1. Glossary: Ice Nucleation Processes and Terminology

Modes of ice nucleation: In this review, we use the terminology proposed by Vali et al. (2015), which we briefly summarize here. *Ice nucleation* is defined as the first appearance of a thermodynamically stable ice phase (Vali et al., 2015). *Homogeneous freezing*, that is, freezing of a supercooled liquid droplet without the presence of a solid particle, is frequently an important process in the upper troposphere, including in the well-studied case of orographic wave clouds. *Heterogeneous ice nucleation* is the occurrence of ice nucleation catalyzed by an ice nucleating particle that enables freezing at a lesser supersaturation or supercooling than would be needed for homogeneous freezing. Several modes of heterogeneous ice nucleation are distinguished, including:

1. *Immersion freezing*, in which freezing initiates on the surface of a particle that is fully immersed in a cloud droplet,
2. *Contact freezing*, in which an interstitial particle collides with a supercooled cloud droplet (Ladino Moreno et al., 2013), and
3. *Deposition ice nucleation*, in which water vapor forms ice directly through deposition onto the surface of a solid particle, without passing through the liquid phase.

In practice, immersion freezing is thought to be the predominant mode of primary, ice formation in mixed-phase clouds, which occur at temperatures too warm for homogeneous freezing (Hoose, Kristjánsson, Chen, & Hazra, 2010). Using large eddy simulations of several idealized and realistic mixed-phase cloud cases, Hande and Hoose (2017) found that immersion mode freezing was the dominant mode of primary ice formation in all cases. This finding is consistent with the physical understanding that ice-nucleating particles typically also are activated efficiently as cloud condensation nuclei, due to their large size, and because they are typically internal mixed with soluble components. Consequently, these particles are likely to be contained inside of cloud droplets, making them available for immersion freezing, but unavailable for contact freezing, which requires the participation of an interstitial particle.

The role of contact freezing in the atmosphere is highly uncertain. Some modeling studies have suggested that contact freezing can contribute to ice formation in mixed-phase clouds for example, Hande and Hoose (2017). The contribution of contact freezing is controlled in part by its relative efficiency; laboratory studies have shown that contact freezing is more efficient than immersion freezing for some particle types and experimental conditions (Fornea et al., 2009; Nagare et al., 2016). However, the contact freezing rate in clouds is limited by the collision rate of interstitial particles with super-cooled liquid droplets. The frequency and efficiency of such collisions in real clouds remains poorly understood at a fundamental level (Kanji et al., 2017; Ladino Moreno et al., 2013), contributing uncertainty to model results aiming to quantify the importance of this freezing mode.

In contrast, the cirrus cloud regime is primarily characterized by a competition between homogeneous freezing, deposition ice nucleation, and immersion freezing. Untangling the relative importance of these mechanisms and their competition has been a long-standing focus of research (DeMott et al., 1997; Kärcher & Voigt, 2006; Sassen & Benson, 2000). We also note that recent literature questions the concept of deposition nucleation as a mechanism of heterogeneous freezing in the cirrus cloud regime. Experimental evidence suggests that nucleation at relative humidities below 100% may instead occur through condensation of a liquid phase in pores, followed by freezing (e.g., David et al., 2019).

There are four main criteria to consider in designing measurements of INPs for the development of aerosol-type-specific parameterizations. Parameterization development efforts should maximize coverage of these four criteria to the best of their ability and clearly state any shortfalls due to experimental limitations.

1. **Temperature and humidity range**: For each relevant aerosol type, ice nucleation activity must be measured over a wide range of temperature and humidity conditions spanning both immersion freezing and deposition nucleation.

Box 2. Tutorial: Direct and Offline Measurements of Ice Nucleating Particles

Historically, **ambient INP measurements** have been obtained using a variety of ice nucleation chamber instrument designs. These instruments can be broadly classified into two categories: direct sampling and offline detection (DeMott, Möhler, et al., 2018).

1. **Direct INP sampling methods** measure the INP efficiency of particles sampled directly from the air by exposing them to the desired supersaturation and temperature profiles, which are achieved by controlling the temperature gradient between two parallel ice coated surfaces (e.g., Kulkarni et al., 2020) or via cooling during simulated adiabatic expansion (e.g., Möhler et al., 2021).
2. **Offline INP detection methods** measure the INP efficiency of particles that have been collected onto filters or other substrates. Samples can be analyzed for deposition ice nucleation by using diffusion chamber ice nucleation experiments, where freezing is observed on collected particles when exposed to controlled freezing conditions (e.g., Budke & Koop, 2015; Murray et al., 2010; Schrod et al., 2016; Tobo, 2016). Immersion freezing can also be measured in cold stage experiments; these experiments are commonly performed by first condensing water onto the collected particles, and then monitoring freezing during controlled cooling (e.g., Wang, Laskin, et al., 2012; Wright & Petters, 2013). In another common offline approach to measure immersion freezing, aqueous suspensions of the collected particles are distributed into well plates, which are monitored for freezing during controlled cooling conditions.

2. **Quantification of ice nucleation events**: The measurement of INPs is sensitive to uncertainties and variability in the number of observed ice nucleation events.
3. **Size and surface area distribution**: The size of the particles must be well-characterized and should cover the entire relevant range.
4. **Aerosol composition**: The aerosol particles that provide major sources of INPs must be chemically characterized. As a minimum requirement, the INPs must be classified by type (dust, sea salt, biological, etc.).

The measurement of temperature and humidity, and its uncertainties, are typically well-characterized (Hiranuma et al., 2015). Therefore, we next summarize challenges and uncertainties associated with the quantification of ice nucleation events, the observed aerosol size distribution, and the measurement of size-resolved particle composition.

2.1. Uncertainties in INP Measurements Associated With Detection, Counting Statistics, and Variability

In INP measurements, ice nucleation events—that is, the formation of the nanoscale initial ice germs—are not detected directly, but after subsequent ice crystal growth, the occurrence of these events are inferred via optical detection of larger ice crystals (see Box 2 for a tutorial on direct and offline measurement methods). Uncertainties in the number of detected ice nucleation events arise from a combination of instrument errors (e.g., the detection efficiency of optical particle counters in real-time instruments) and the underlying stochasticity of ice formation. Most studies report an upper limit for the combined relative uncertainty of frozen fractions, typically between 10% and 30% (Hiranuma et al., 2015). The contribution of stochasticity to the combined uncertainty can be substantially higher when sample sizes are low, for example, limited to the number of wells in a freezing assay (Alpert & Knopf, 2016). Additionally, the stochasticity of freezing produces a dependence of the apparent J_{het} or n_s value on experimental design such as the experimental cooling rate (Knopf et al., 2020) and the duration of exposure to freezing temperatures and humidities (Herbert et al., 2014). Experiments that report results only in the form of deterministic parameterizations typically neglect the impacts of time dependence, since these effects are usually much smaller than temperature dependence for atmospherically relevant situations, as discussed above (Ervens & Feingold, 2013). However, since accounting for this time-dependent behavior can resolve some discrepancies between different experimental systems (Herbert et al., 2014), recent theoretical studies recommend either accounting for time-dependent behavior explicitly (Knopf et al., 2020) or normalizing to remove its effects (Herbert et al., 2014).

In addition, since INP concentrations in the ambient atmosphere are frequently lognormally distributed, researchers should consider carefully the most appropriate methods of quantifying variability in INP counts. As a best practice, the geometric standard deviation should be reported alongside, or in lieu of, the standard deviation for lognormally distributed variables (Limpert et al., 2008).

2.2. Uncertainties Associated With the Aerosol Size Distribution

Another important source of uncertainty associated with all modes of ice nucleation, which is often the most challenging to quantify, is the aerosol size distribution, which determines the total available aerosol surface area and therefore influences the probability of ice nucleation events. An accurate measurement of the surface area of the accumulation and coarse mode aerosol in the relevant size range (e.g., from tens of nm to 10 μm) is therefore required to accurately calculate the coefficient n_s or J_{het}.

For droplet freezing measurements of bulk material, the ice nucleation surface area is typically calculated as a function of the mass concentration of particulate sample that has been diluted in water and then distributed among droplets or wells. The mass per droplet or well is multiplied by the specific surface area (m^2/g) as determined from gas adsorption (Brunauer-Emmett-Teller or BET approach) to obtain the surface area per well or droplet. The choice of gas used for adsorption may represent a source of uncertainty in this approach as gases may differ in their interactions with particle surfaces. Ice nucleation studies have used nitrogen, water vapor and argon gas to quantify the specific surface area (Hiranuma et al., 2015; Umo et al., 2021). While Hiranuma et al. (2015) found only minor differences between the specific surface areas of illite samples measured using nitrogen and water vapor, other aerosol types may exhibit different behavior due to differences in physicochemical surface properties and morphology. An important limitation of this method is that it neglects the sedimentation of suspended particles that can potentially occur over the course of a measurement period, leading to a potential underestimation of the freezing rate.

Additionally, deviations from an idealized, single-component, monodisperse aerosol can produce nonideal behaviors in INP quantification experiments. In particular, in offline freezing experiments there is a potential for large variations in the INP surface area present in each well or droplet, which Alpert and Knopf (2016) showed could lead to both systematic errors and a substantial broadening of the uncertainty ranges in the inferred values of J_{het}. Since freezing probability is determined by both particle surface area and particle surface freezing efficiency, particle-to-particle differences in freezing efficiency could cause similar behavior (Murray et al., 2012). These deviations from ideality are frequently neglected as a first-order approximation due to the difficulty of characterizing the spread in freezing efficiency among a population of particles.

For experiments with dry-dispersed aerosol samples, aerosol surface area has been measured using either BET analysis or geometric surface areas derived from aerosol sizing instruments including the scanning mobility particle sizer (SMPS) and aerodynamic particle sizer (APS; Hiranuma et al., 2015). Uncertainties can also arise from other instrumental nonidealities (e.g., particle losses or exposure of particles to varied supersaturation), which are specific to the experimental conditions as well as particle size and aerosol type (DeMott et al., 2015; Garimella et al., 2017) and thus, in most cases, not all explicitly quantified. A related source of uncertainty is particle shape: many INPs (e.g., dust particles) are aspherical and have size-dependent shape and porosity (Alexander et al., 2016). Aspherical particles have different surface areas and may experience different instrument losses, compared to spherical particles with identical equivalent aerodynamic diameters. These effects are not captured by bulk sizing methods such as the SMPS and APS.

Particle losses by design – due to an impactor or unavoidable losses to surfaces of inlets, tubing, and other components – often lead to a systematic undersampling of larger (supermicron) particles, which have been observed to contribute disproportionately to ice nucleation efficiencies in some environments (Mason et al., 2016; Suski, Hill, et al., 2018), and are therefore currently understudied. We also emphasize that it is important to consider the particle size range quantified in sizing instruments compared to the particle size range measured for INP analysis, truncating size distribution measurements, if applicable.

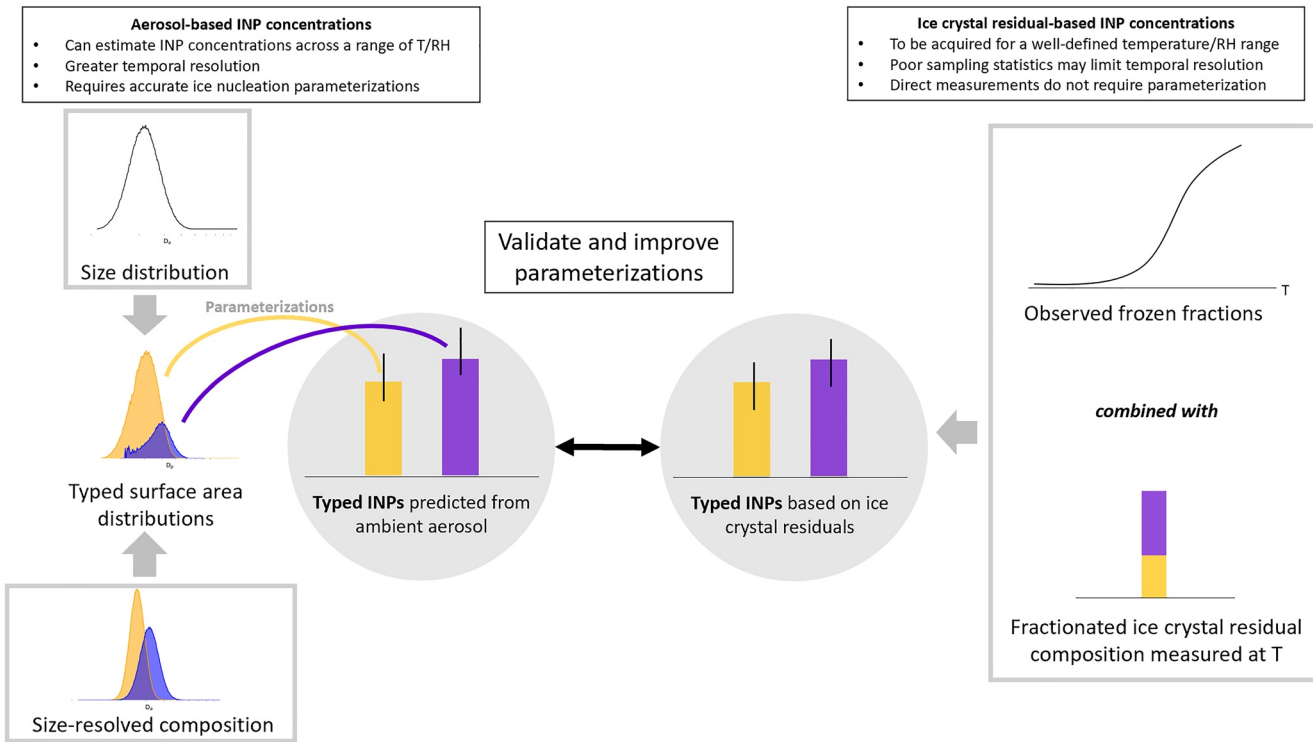

Figure 2. Schematic for how measurements can be used to determine the concentrations of ice-nucleating particles (INPs) arising from different particle types, for the purpose of closure calculations in environments with complex aerosol compositions. The left side of the figure shows the aerosol measurement-based approach, where measurements of aerosol size and composition are used to calculate particle surface area distributions for each relevant particle type, and type-dependent parameterizations of INP efficiency are applied to estimate INP concentrations arising from each particle type. Observational closure is achieved when the total number of INPs calculated by this method agrees with an independent measurement of INP number (as a function of temperature and humidity conditions). The right side shows the ice crystal residual approach, where the composition of INPs that are activated into ice crystals is measured directly. A quantitative measurement of type-dependent INP concentrations at a specific temperature can then be obtained by scaling the INP concentrations by the type-dependent fraction of INP residuals. This measurement, while challenging, can enable a stronger test of process-level understanding of INP concentrations, by enabling a type-dependent closure calculation. In a type-dependent closure, the number of INPs for each aerosol type would be calculated and compared between the aerosol measurement-based approach, and the ice crystal residual approach.

2.3. Size-Resolved Composition Measurements for INP Closure Analysis

Since ice nucleation activity of aerosols (whether expressed as J_{het} or n_s) varies dramatically between aerosol types (Kanji et al., 2017; Murray et al., 2012), adequate predictive understanding of INPs ultimately require the use of aerosol-type-dependent INP parameterizations. While field studies correlating INP concentrations with particle types have led to insights about the dominant sources of INP at specific locations and times (e.g., Huffman et al., 2013; Mason et al., 2015; Si et al., 2019), there is also a need for more experiments that provide independent, quantitative tests for our understanding of the processes driving INP variability in the atmosphere. A stronger and more complete test of predictability is an experiment that directly evaluates whether INP parameterizations produce accurate simulations of INP numbers in real-world ambient measurements, termed a "closure" experiment (Knopf et al., 2021).

As discussed previously, particle size, composition, and surface features can all impact the ice-nucleating activity of particles. In a prototypical closure experiment, as illustrated in Figure 2, size-resolved aerosol composition is measured and used as an input to parameterizations to calculate the expected number of INPs. This predicted INP number is then compared with the observed INP number. Closure is achieved if the predicted and observed INP numbers agree to within their respective uncertainties. If the experiment also includes a measurement of size-resolved composition of ice crystal residuals, the closure analysis can be conducted separately for each particle type, providing additional insight into sources of measurement discrepancies.

Within this framework, an ideal suite of complementary particle characterization techniques that could be used for INP closure and attribution studies, as well as to produce additional insight into physical processes, would include the following capabilities:

1. Simultaneously measure size and composition of ambient particles on a single-particle basis (required input for closure calculations).
2. Distinguish between known classes of INPs, for example, dust, sea spray, bioparticles, biomass burning (required input for closure calculations).
3. Simultaneously measure size and composition of INPs on a single-particle basis (valuable for verification of closure calculations).
4. Characterize surface morphology and chemical features of INPs (desirable to provide insight into potential reasons for gaps in closure).
5. Characterize any soluble components of the aerosol, the presence of which can modify freezing temperatures.

No single instrument or method is currently able to observe all of these quantities and the rarity of INPs makes it extremely challenging to separate them from the far more numerous interstitial particles and droplets. Additionally, many of the widely available methods that are useful for studies of cloud condensation nuclei (CCN) and aerosol optical properties are of limited utility for INP closure studies. As an example, the most commonly used instrument for measuring composition, the Aerosol Mass Spectrometer, provides information on bulk aerosol composition of non-refractory aerosol and is unable to characterize refractory particles such as dust or sea spray, making it unsuitable for INP closure studies. Therefore, in the following section we discuss in greater detail some of the available methods that are able to characterize the size-resolved composition of atmospheric particles and are particularly valuable for INP closure studies.

2.3.1. Size-Resolved Composition Measurements of Atmospheric Particles

Since INP parameterizations depend on both particle surface area and aerosol composition (via the type-dependent ice nucleation efficiency, J_{het} and n_s), a size-resolved composition measurement is critical to INP closure studies. Size-resolved composition measurements capable of distinguishing major INP classes can be achieved through either real-time or offline approaches, with important tradeoffs. Real-time approaches offer the potential to discern changes in sources during the diurnal cycle or as air masses change, while the sampling times required by offline approaches may at times be too long to detect sub-daily variability. However, real-time approaches historically have achieved only small sample sizes and offer less-detailed insight into particle morphology, surface characteristics, and mixing state.

Real-time composition measurements that are capable of distinguishing all relevant aerosol types, including dust, sea spray, biological particles, and elemental and organic carbon, as a function of particle size, with high temporal resolution are extremely challenging. Most commonly, single-particle mass spectrometers are used to gain information on the real-time, size-resolved composition of many of the relevant particle classes (sea spray, dust, and biological particles). The results from these instruments are usually not directly comparable because they are custom-built instruments that employ inlets with different transmission efficiencies; different detection or ionizing lasers, which affects size-range sensitivity to aerosol composition, respectively; and different mass spectral analyzers (see Zawadowicz et al. [2020] for some discussion of this). However, particles analyzed by different SPMS designs often produce similar sets of ion markers that are distinguishing or characteristic of specific INP types. Therefore, studies produced using one instrument can still be informative for interpreting and anticipating results from other SPMS instruments with different designs, and for studies aimed at understanding the properties of different types of INPs.

Offline measurements of size-resolved composition can be achieved by using a multistage impactor such as the Microorifice Uniform Deposit Impactor (or MOUDI) to collect particle samples in different size ranges, and subsequently analyzing these samples using multimodal microscopy-spectroscopy approaches (Laskin et al., 2016, Figure 3). Alternatively, automated computer-controlled electron microscopy systems are now capable of simultaneously classifying and sizing large sample sizes of particles collected on filters or other substrates. Relative to real-time measurement approaches, these offline methods have far lower temporal resolution, with typical sampling times in the range of 30 min to a few hours depending on the planned analysis. However, offline methods have the advantage that they are able to access a broader size range, including those >2.5 μm, a bound used by some real-time measurements to assist in distinguishing small ice particles from aerosol, and can provide

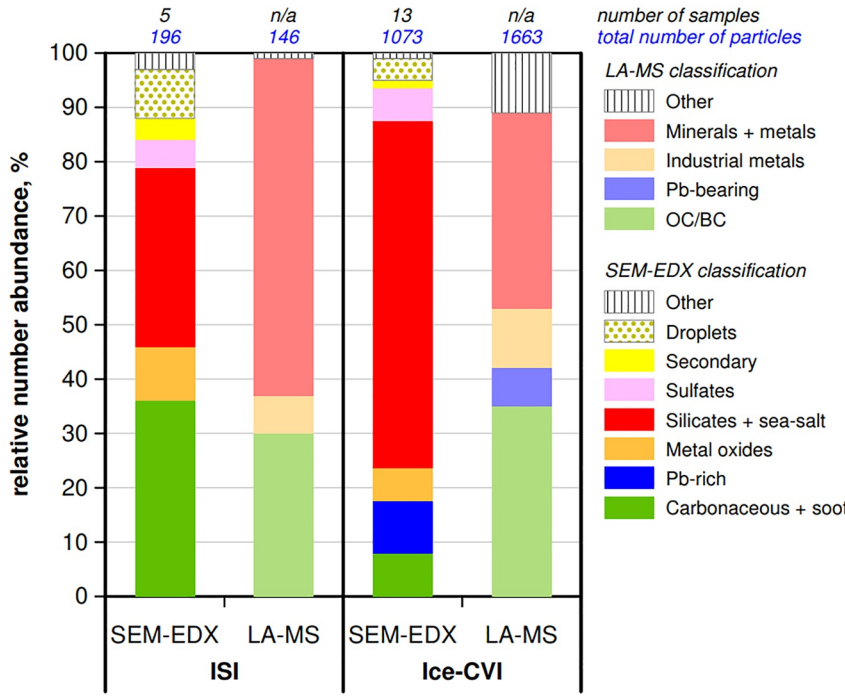

Figure 3. Example of chemical characterization of ice residuals separated by Ice Selective Inlet and Ice Counterflow Virtual Impactor from atmospheric mixed-phase cloud at the high alpine research station at Jungfraujoch. Chemical characterization was performed both offline by scanning electron microscopy coupled with energy dispersive X-ray spectroscopy, and online by laser ablation mass spectrometry. Reproduced from Worringen et al. (2015).

unique insights (e.g., molecular-level characterization of particle surfaces). Offline techniques will be discussed in greater detail in Section 2.3.3 and real-time methods in Section 2.3.4.

2.3.2. Separation of Immersion Freezing INPs for Direct Chemical Analysis

When available, direct chemical analysis of INPs can provide additional insight into the identity of these particles. Direct chemical analysis can be accomplished by nucleating immersed particles into ice crystals within a continuous flow diffusion chamber (CFDC)-type instrument or a cloud chamber (Möhler et al., 2001) and isolating nucleated ice crystals by taking advantage of their relatively large size and inertia. Ice crystals containing INPs may be either (a) collected via impaction onto substrates for offline microscopy analysis (e.g., Kreidenweis et al., 1998, Figure 3) or (b) preferentially sampled via counterflow virtual impaction and analyzed in situ with single-particle mass spectrometry (Cziczo et al., 2003; Hiranuma et al., 2016; Suski, Bell, et al., 2018). A similar approach can be used to analyze the residual material contained in cloud ice from for example, aircraft platforms (e.g., Cziczo et al., 2017; Noone et al., 1988; Ogren et al., 1985; Twohy et al., 1997; Zelenyuk et al., 2015).

Ice crystal impaction onto substrates and subsequent offline analysis of residuals were first demonstrated by Kreidenweis et al. (1998), and have since been used to determine the fractional contribution of different particle types to INPs present in agricultural soils (Suski, Hill, et al., 2018; Tobo et al., 2014), boundary layer and free troposphere (Chen et al., 1998; Prenni, DeMott, et al., 2007; Prenni, Demott, et al., 2009; Richardson et al., 2007; Rogers et al., 2001; Twohy et al., 2021), biomass burning particles (Barry et al., 2021; McCluskey et al., 2014), and nascent sea spray (McCluskey, Hill, Sultana, et al., 2018). INPs have been characterized using real-time methods specialized for the detection of individual particle classes such as refractory black carbon (single particle soot photometer, SP2; Crawford et al., 2011), fluorescent bioparticles (ultraviolet APS; Huffman et al., 2013), and wideband integrated bioaerosol sensor (Boose, Sierau, et al., 2016; Gosselin et al., 2016). Additionally, studies have utilized sampling techniques that remove targeted material upstream of an offline INP measurement, for example, using a heating tube to remove heat labile material (Suski, Hill, et al., 2018) or an SP2 to remove refractory black carbon (Levin et al., 2014) and measuring the impact of the targeted material on the INP number concentration. However, these specialized methods cannot distinguish all major classes of INPs.

An important limitation of studies that chemically analyze INPs arises from their reliance on a counterflow virtual impactor (CVI) or a pumped counterflow virtual impactor (PCVI) to aerodynamically separate the ice crystals formed from INPs active at the measurement temperature from interstitial particles and/or liquid droplets. The operating principle of the PCVI is that the airstream containing sampled particles is confronted with a counterflow of particulate-free air (Boulter et al., 2006). Larger particles (i.e., activated ice crystals and cloud droplets) are able to penetrate the counterflow due to their greater inertia, while smaller particles (i.e., interstitial, non-INPs) are deflected and excluded from the downstream sample. The median cutoff size separating the transmitted and excluded particles is a function of the incoming flow rate and the counterflow rate, and can be adjusted by changing these flows (Pekour & Cziczo, 2011).

However, the PCVI imperfectly separates ice crystals from interstitial particles: smaller interstitial particles can pass through the PCVI in the wake of larger particles or ice crystals (Pekour & Cziczo, 2011). Additionally, a small number of the much more abundant particles below the PCVI's cutoff diameter are not deflected by the counterflow (Boulter et al., 2006; Kulkarni et al., 2011). Both of these effects can lead to the transmission of non-INPs through the PCVI, creating a measurement artifact. For example, Boulter et al. (2006) reports that when the PCVI was configured to have "a 50% cutoff aerodynamic diameter of ca. 1.50 μm, particles less than 1 μm in diameter were transmitted with an efficiency of less than 0.002%." Under ambient conditions, however, it is common for the concentrations of submicron particles to exceed those of supermicron particles by two or more orders of magnitude, with the consequence that breakthrough transmission events can be a confounding factor in such experiments. Due to the rarity of INPs, this imperfect separation by the PCVI can potentially result in non-INPs outnumbering INPs among the transmitted particles during ambient sampling. This is a central challenge of experiments that rely on residual separation and effectively establishes a lower limit on the INP concentrations, as a fraction of the total aerosol number concentration, for which residuals can be meaningfully separated and chemically characterized.

2.3.3. Laboratory Measurements of Particle Surface Chemistry and Morphology

Because heterogeneous ice nucleation is fundamentally a process that occurs on particle surfaces, surface topological features (Friddle & Thürmer, 2020) and variations in surface composition have an effect on the observed ice nucleation properties. Achieving greater insight into the relationship between particle surface properties and their ice nucleation activity can improve understanding of how the differences in ice-nucleating efficiency between different material arise. This deeper understanding has the potential to enable further insights into the conditions under which existing parameterizations may fail in their predictive validity, for example, in response to chemical transformations in the atmosphere. Additionally, laboratory measurements can improve understanding of which subcomponents of complex atmospheric particles (e.g., soot or mixed dust-biological particles) are primarily responsible for controlling their activity as INPs. At present, however, more work is needed to develop and evaluate aerosol-specific parameterizations based on particle composition before parameterizations integrating the impact of surface features can be explored for use in large-scale models.

Several analytical tools can provide insights into the surface chemistry and morphology of INPs at either the single particle or bulk aerosol level. SEM/EDX can provide detailed surface characterization, morphology and elemental composition of individual particles, while transmission electron microscopy (TEM) provides detailed internal structures of particles with spatial resolution at the atomic scale (e.g., Figure 4; China et al., 2017). Additionally, atomic force microscopy can provide images of the topography of individual particles (X. Woodward et al., 2015), and has been deployed to characterize individual INPs in at least one study (Iwata & Matsuki, 2018).

Scanning transmission X-ray microscopy coupled with near-edge X-ray absorption fine structure spectroscopy (STXM/NEXAFS) provides chemical bonding, oxidation state, and information about different functional groups present in individual particles at high spatial resolution (~30 nm). STXM/NEXAFS has been deployed to understand the nature of organic matter and the mixing state of individual INPs (Knopf et al., 2014; Wang et al., 2016, Figure 5), and Raman spectroscopy has been used to study the spatial distribution of chemicals within ambient aerosols and the ice active fraction of those aerosols (Baustian et al., 2012; Iwata & Matsuki, 2018).

Cold-stage ice nucleation experiments involve exposing particles on a flat substrate to controlled freezing conditions and observing freezing events, often with automated detection by an optical camera or microscope (e.g., Budke & Koop, 2015; Murray et al., 2010, 2012; Tobo, 2016). Cold-stage experiments can then be complemented by offline chemical and physical analysis of particles that initiated freezing. A more recent development

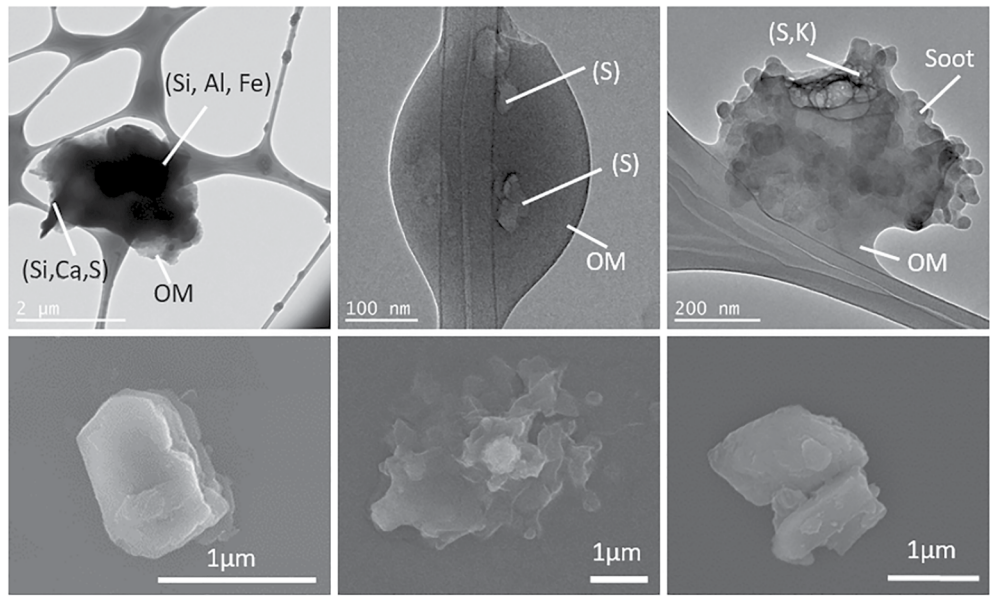

Figure 4. Prototypical measurement of the morphology and chemical composition aerosols and ice-nucleating particles (INPs), for a sample collected an alpine site in the Azores, Portugal. Top row: transmission electron microscopy images of an aerosol population. Bottom row: SEM images of INPs. Reproduced from China et al. (2017).

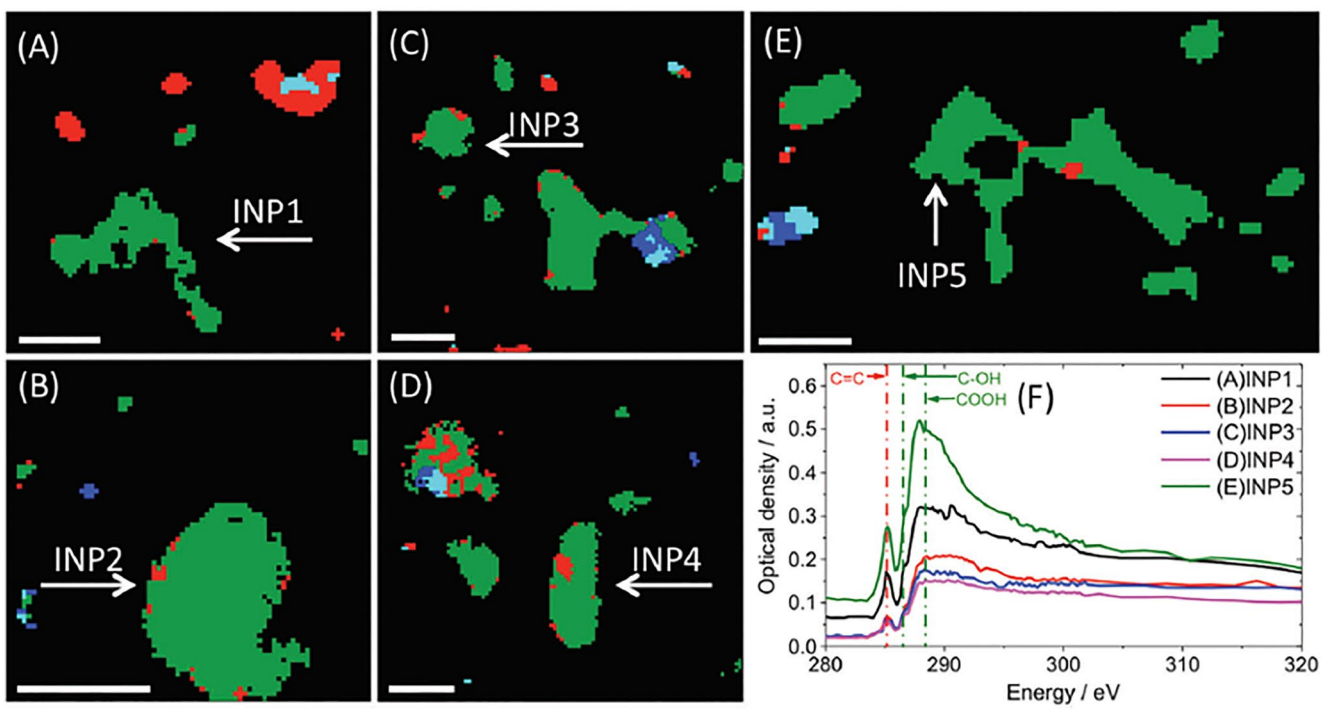

Figure 5. Example of chemical imaging of ice-nucleating particles (INPs) using IN-ESEM and STXM/NEXAFS on particles collected at California during the California Research at the Nexus of Air Quality and Climate Change (CalNex) field campaign in 2010. Sublimated ice crystals were tracked to identify INPs in IN-ESEM and later probed using STXM/NEXAFS for chemical imaging. Different colors in the compositional maps (a–e) represent organic carbon (green), organics and inorganics (blue), organics and elemental carbon (red), and a mixture of organic, elemental carbon, and inorganics (cyan). Panel (f) shows the carbon K-edge spectra for the five identified INPs (as indicated by arrows in a–e). Scale bars are 1 mm. Reproduced from B. Wang et al. (2016).

is an ice nucleation stage interfaced with an environmental scanning electron microscope, which is capable of directly observing ice formation on individual particles with high spatial resolution, providing new insights into the initiation stages of ice formation and its relationship to specific particle surface sites (Lybrand et al., 2021; Wang et al., 2016).

Other analytical methods such as helium ion microscopy (Wang et al., 2016), X-ray photoelectron spectroscopy (Kanji et al., 2008; J. Yun et al., 2020), time-of-flight secondary ion mass spectrometry and NanoSIMS (China et al., 2018; Harris et al., 2013; Peterson & Tyler, 2003; Sobanska et al., 2014) can provide detailed insights into surface topography and chemistry, but to the best of our knowledge have not yet been extensively deployed for ice nucleation research, indicating that significant unexplored frontiers still exist in this area of research.

2.3.4. Real-Time Characterization of Particles and INPs With Single-Particle Mass Spectrometry

As discussed above, single-particle mass spectrometry (SPMS) is the only real-time method currently capable of distinguishing all major INP classes (dust, sea spray, and biological particles). SPMS is also currently the only real-time method capable of chemically classifying particles with sufficient sensitivity to directly characterize INPs that have been separated from non-activated particles and cloud droplets using a PCVI or ice-selecting pumped counterflow virtual impactor (IS-PCVI). SPMS measures the size-resolved composition of individual particles and is well-suited for residual characterization because of its high sensitivity and temporal resolution. SPMS has been applied to characterize aerosol particles and INP sources in the free troposphere (Baustian et al., 2012; DeMott, Cziczo, et al., 2003; Richardson et al., 2007), including onboard aircraft sampling ice crystal residuals through the CVI inlet (Zelenyuk et al., 2015), in urban settings (Corbin et al., 2012), and in the marine boundary layer (Cornwell et al., 2019). When referenced to an independent particle-size distribution measurement, SPMS methods can produce quantitative estimates of the available aerosol surface area (Cziczo et al., 2009; Froyd et al., 2019; Qin et al., 2006; Zawadowicz et al., 2015). A detailed description of different SPMS instruments and methods, including their relative sensitivities to different aerosol types and/or sizes, can be found in dedicated reviews (e.g., Murphy, 2007; Nash et al., 2006; Passig & Zimmermann, 2021; Pratt & Prather, 2012).

In a recent study, Cornwell, McCluskey, et al. (2021) demonstrated a novel method for using integrated CFDC-SPMS measurements to directly derive heterogeneous immersion freezing rate coefficients for two different classes of ambient aerosols, dust and sea spray, in a mixed air mass containing both types of particles. This derivation is performed by separately calculating the frozen fraction for droplets containing each particle type; in a mixed air mass this is only possible when INPs have been separated and chemically classified using a CFDC-SPMS experiment. Residual characterization experiments therefore offer an unparalleled ability to characterize ice nucleation properties directly for ambient aerosol. In this way, CFDC-SPMS experiments provide an important complement to laboratory-based studies of INPs, which require the use of laboratory-generated aerosol samples that might not perfectly reproduce the ambient aerosol.

Despite the obvious advantages of an integrated CFDC-SPMS for real-time direct characterization of INPs, an important limitation of this approach has been the small sample sizes that were historically achievable in field experiments. This challenge is illustrated by Figure 6, which shows the sampling time that would be required to achieve different sample sizes for a prototypical integrated CFDC-SPMS experiment. For instance, to chemically characterize 100 ice crystal residuals at an ambient INP concentration of 10 L^{-1} would require approximately 50 hr of sampling. In field studies with dynamically changing meteorology and atmospheric sources it is frequently difficult to sample the same air mass for sufficient duration to achieve a statistically significant comparison with a contrasting air mass.

Recent studies have demonstrated that INP sample sizes can be increased by deploying an aerosol aerodynamic concentrator upstream of the CFDC (Boose, Kanji, et al., 2016; Cornwell et al., 2019; Suski, Hill, et al., 2018). Aerosol concentrators work on the principle of virtual impaction and consequently preferentially enhance the concentrations of larger particles (Gute et al., 2019; Romay et al., 2002). This property makes them well-suited for enhancing INP concentrations due to the strong relationship between particle surface area and ice-nucleating activity.

Given the low atmospheric concentrations of INPs, it can be difficult to acquire sufficiently large samples for robust INP classification into different aerosol types. Coatings and inclusions can also significantly impact how a particle nucleates ice (Kanji et al., 2019), but their presence is difficult to quantify using SPMS. Robust

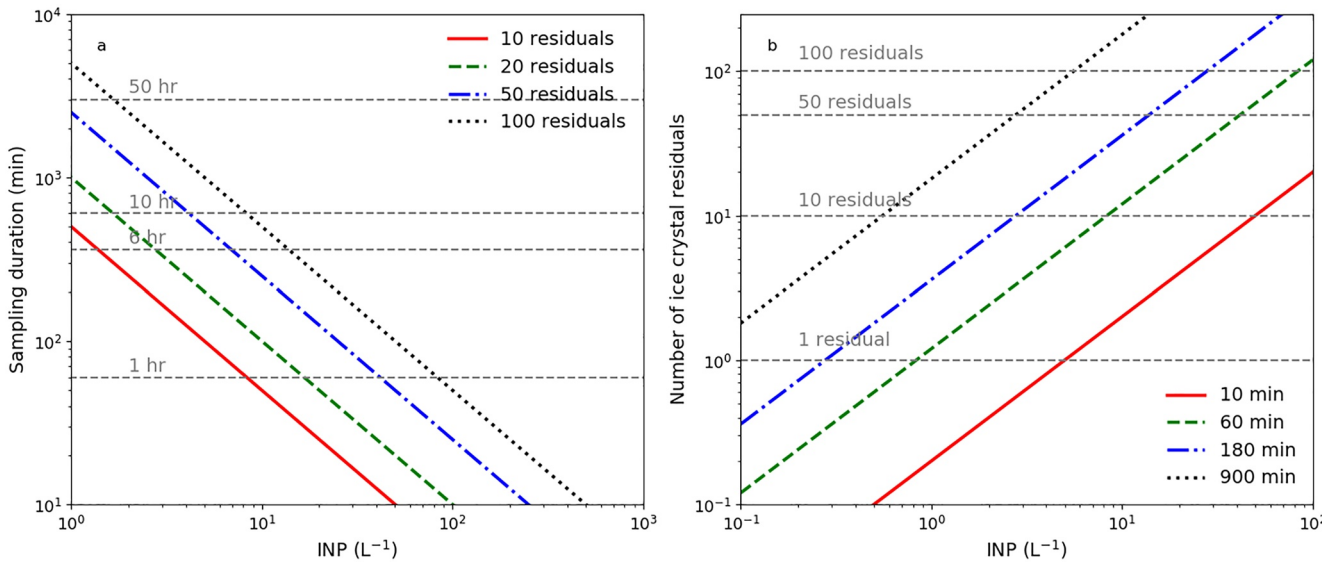

Figure 6. Illustration of the challenges faced in achieving statistically significant samples with a prototypical integrated CFDC-SPMS measurement: (a) shows the sampling time required to measure a certain number of residuals as a function of the ambient ice-nucleating particle (INP) concentration and (b) shows the number of particles estimated to be measured given a particular sample period duration as a function of ambient INP concentration. The number of ice crystal residuals chemically characterized by the SMPS, N_{ICR}, can be calculated as $N_{ICR} = t \times Q \times N_{INP} \times TE_{PCVI} \times HR_{SPMS}$, where t [min] is the sampling duration, Q [L min^{-1}] is the sample flow rate of the SPMS, TE_{PCVI} is the transmission efficiency for ice crystals through the PCVI to the SPMS [a scalar value between 0 and 1], and HR_{SPMS} is the hit (or detection) rate of the SPMS, that is, the fraction of particles entering the SPMS that are successfully detected and chemically characterized [a scalar value between 0 and 1]. The estimates shown here use the following first-order assumptions for these parameters: $Q = 0.1$ L min^{-1}, $TE_{PCVI} = 0.2$, and $HR_{SPMS} = 0.2$. Additionally, particle losses to sampling inefficiencies are neglected.

benchmarking of SPMS spectra with laboratory analogs of relevant particle types is therefore important to improve and build confidence in particle classification schemes.

2.4. Impacts of Soluble Components on Freezing Temperatures

Since INPs in ambient air are typically internally mixed with soluble components, the freezing temperature of ambient immersion-mode INPs could be modified by the presence of such solutes. In most atmospheric ice nucleation research, dilute solutes have typically been assumed to suppress freezing, to a small degree that can be neglected for most practical applications in the atmosphere. Additionally, research addressing solute impacts on atmospheric ice nucleation has often assumed that droplets can be adequately approximated as ideal solutions, for example, by using a water activity-based approach (Knopf & Alpert, 2013). Some recent studies, however, complicate this understanding. For example, Whale et al. (2018) summarized previous literature and reported new results showing that dilute solutions of various constituents either enhanced or suppressed freezing temperatures, but the effects differed for different pairings of ice nucleators and solutes. This is in contrast to the idealized understanding, suggesting that complex molecular-level interactions are impacting the ice nucleation process in some systems.

3. Predictive Understanding of INPs for Atmospheric Modeling

While observations provide information at a particular location and time, numerical models enable studies that investigate how INPs may influence and be influenced by surrounding processes across various temporal and spatial scales. The value of modeling tools for understanding INP interactions with the Earth system has been demonstrated in many previous studies. For example, models have been used to investigate the significance of dust mineralogy for global ice nucleation (Atkinson et al., 2013), the relevance of sea spray aerosol as a source of immersion INPs in the remote marine boundary layer (Burrows, Hoose, et al., 2013; McCluskey et al., 2019; Vergara-Temprado et al., 2017; Wilson et al., 2015), and the relevance of biogenic aerosols for shallow cloud layers in Northern Europe (O'Sullivan et al., 2018). Models have also been used to study the impacts of changes

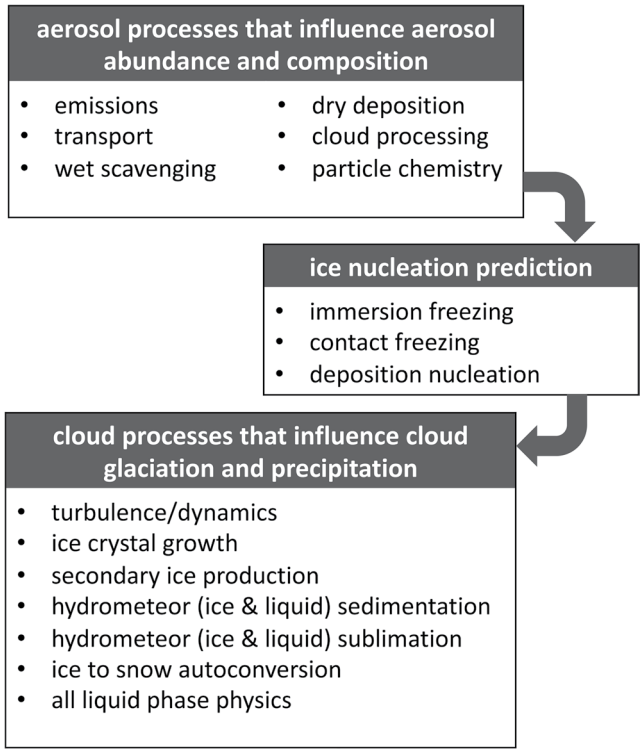

Figure 7. Overview of the aerosol processes that would be required for a comprehensive treatment of ice-nucleating particles (INPs) prediction in atmospheric models (top), and the cloud processes required for a comprehensive treatment of cloud responses to INPs. The large number of relevant processes, in combination with the multiple spatial and temporal scales inherently required for simulation of these processes, contribute to the challenges associated with using models to understand INP impacts on clouds and climate.

in anthropogenic emissions of particles and their precursor gases, such as reductions in INP efficiency due to coating of particles by condensed gases (Kulkarni et al., 2014).

In addition to these focused studies, the implementation of aerosol-aware INP schemes in regional and global climate and Earth system models provides a pathway toward understanding how atmospheric INPs may have changed in the past (Sagoo & Storelvmo, 2017) or may change in the future due to changes in land use, land cover, and climate (Webb & Pierre, 2018). Improved modeling capabilities will then enable greater insights into how these changes can impact precipitation, cloud radiative effects, and cloud-climate feedbacks. Consequently, in the past 10–15 years, the modeling community has increasingly begun to shift away from representations of INPs that depend only on temperature and humidity, and toward aerosol-aware representations of cloud droplet freezing in regional and global models. Importantly, using aerosol-aware cloud ice schemes means that models are increasingly subject to errors and uncertainties in the simulation of INPs. In turn, the simulation of INPs is subject to the limitations of present-day models in representing aerosol and cloud processes. The interplay between uncertainties in simulation of aerosol, INP, and cloud processes is conceptually represented in (Figure 7).

Efforts to link ice nucleation to aerosols began evolving in the early 2000s when parameterizations of heterogeneous freezing were developed and implemented in the European Center for Medium Range Weather Forecast General Circulation Model (GCM; Lohmann et al., 2004). These treatments of heterogeneous freezing by dust and homogeneous freezing by sulfate aerosol for cirrus clouds developed by Liu and Penner (2005) were subsequently incorporated into the National Center for Atmospheric Research Community Atmospheric Model (CAM; Eidhammer et al., 2017; Gettelman et al., 2010; Liu et al., 2007). Most of these models consider homogeneous freezing of sulfate aerosols and heterogeneous immersion freezing of dust aerosols. The NASA GEOS-5 global model predicts heterogeneous ice nucleation using a parameterization originally developed by Phillips et al. (2013) and implemented by Barahona et al. (2014). This parameterization includes immersion and deposition ice nucleation on dust, black carbon, and soluble organic aerosols. Hoose, Kristjánsson, Chen, and Hazra (2010) implemented a CNT-based parameterization of heterogeneous ice nucleation for mineral dust, soot, and biological particles in the CAM-Oslo global climate model.

Another important consideration is that the scale of a modeling framework must be appropriately matched to the scale of processes to be studied. This is a challenge for simulation of INPs and their interactions in the Earth system, since the relevant aerosol and cloud processes occur across multiple scales. With simulations over limited spatial domains (e.g., tens of kilometers) and short time periods (e.g., a single day to several days), Large Eddy Simulations (LES) are best suited to resolve the relevant cloud processes for investigating ice nucleation. However, LES cannot address questions related to the sources and removal of INPs at regional and global scales, global-scale radiative impacts, or climate feedbacks. Meanwhile, regional and global models are a useful tool to study cloud-climate feedbacks and large-scale aerosol transport. However, large-scale models lack the temporal and spatial resolution required to simulate interactions between aerosol chemistry and physics, cloud microphysics, cloud dynamics, and the cloud environment. In fact, a recent intercomparison showed that no global model correctly simulated the regional and seasonal patterns in cloud phase as observed by lidar (Komurcu et al., 2014). So on fundamental grounds and due to poor agreement with observational constraints we have low confidence that today's regional and global models can adequately simulate every process controlling the sensitivity of clouds and climate to INPs.

With these caveats in mind, it is desirable to target available resources toward measurements and model developments that will result in the greatest improvements in modeling of cloud phase, radiative properties, and precipitation. Here we describe the current practices and challenges in four areas that are needed to advance predictive

understanding of INPs: (a) implementation of INP parameterizations (Section 3.1); (b) simulation of aerosols that contribute to INPs (Section 3.2); (c) constraining INP simulations and evaluating model predictive skill (Section 3.3); and (d) using models to understand how and when INPs affect clouds (Section 3.4).

3.1. Implementation of INP Parameterizations in Models

While a variety of INP parameterizations have been developed on the basis of laboratory and field measurements in recent decades, modelers still frequently face challenges in selecting and implementing an appropriate approach for representing INPs and the particles that provide them. This challenge is particularly pronounced in regional- and global-scale simulations, where a dynamic simulation of particle sources is desirable, and where cloud processes are not fully resolved. Here, we discuss three challenges in particular: (a) the selection of parameterizations; (b) treatment of gaps in parameterized space; and (c) representativeness of parameterizations for regional or global application.

3.1.1. Selection of Parameterizations: Time-Dependent or Deterministic Parameterizations and the Shift Toward Aerosol-Aware and Type-Dependent INP Parameterizations

As outlined in Section 2, INP parameterizations developed from experimental data can be divided into two categories: time-dependent and deterministic parameterizations. As models have increasingly moved from solely temperature-dependent to aerosol-aware INP representations, different modeling groups have made different choices about which parameterization approach to incorporate. Additionally, some models employ different parameterization approaches to represent different modes of freezing and sources of freezing nuclei; for example, contact, immersion, and deposition freezing may be handled using a mix of deterministic and CNT-based parameterizations, with varying degrees of empirical support available for these parameterizations, depending on the particle type.

Time-dependent parameterizations based on CNT are employed in a number of widely used atmospheric models: examples include the CAM6, E3SM, and CAM-Oslo models at the climate modeling scale (Hoose, Kristjánsson, Chen, & Hazra, 2010; Wang et al., 2014); CNT has also been employed at cloud-resolving scales (e.g., Khvorostyanov & Curry, 2004, 2005) and in parcel models (Ervens & Feingold, 2013; Herbert et al., 2015). By including a representation of time-dependent behavior, CNT schemes arguably can capture more aspects of physical freezing behavior. However, it is unclear that a representation of the time dependence of freezing is required for modeling INP impacts on clouds. In most atmospherically relevant situations, nearly all freezing occurs within the first seconds; this is far more rapid than the typical time steps of regional and global models, which are on the order of minutes to tens of minutes. For example, Ervens and Feingold (2013) examined the relevance of time dependence for simulating immersion freezing using cloud parcel model ensembles. This study found that simulated cloud responses were far less sensitive to time dependence than to cloud drop temperature, INP diameter, and particle type (which controlled particle efficacy as INP). They concluded that freezing could be sufficiently approximated by a deterministic formulation in large-scale models. For this reason, and because deterministic schemes are computationally efficient and easy to implement, model developers frequently elect to use the deterministic approximation of ice nucleation.

Some models use a mixture of parameterization approaches: for example, Liu et al. (2007) introduced the Liu and Penner (2005) parameterization into CAM3, which treats the competition between homogeneous nucleation of sulfate and heterogeneous immersion nucleation on soot in cirrus. In this scheme, the treatment of homogeneous nucleation was based on CNT, while the treatment of heterogeneous nucleation was deterministic. Additionally, in the CAM3 implementation described in Liu et al. (2007), the temperature-dependent Young (1974) parameterization was used to treat contact freezing in mixed-phase clouds, while the humidity-dependent parameterization of Meyers et al. (1992) was assumed to represent deposition/condensation freezing on mineral dust.

More recently, some modeling studies have implemented the field-derived parameterization of DeMott et al. (2010), or D10, which uses an empirical relationship between the number of larger particles (with diameters greater than 0.5 μm) and the number of immersion freezing INPs, as a function of temperature. This parameterization has been applied in simulations both in global models (Miltenberger et al., 2018; Solomon et al., 2015; Storelvmo et al., 2011; Tan & Storelvmo, 2019; Xie et al., 2013) and in limited-domain cloud-resolving models (Hawker et al., 2021; Saleeby & van den Heever, 2013; Sarangi et al., 2018; Sullivan et al., 2018; Y. Yun et al., 2020). D10 is empirically derived from ambient measurements and so does not suffer from concerns about

the representativeness of laboratory samples. Additionally, D10 does not require detailed particle properties to be represented, which facilitates ease of use in some models. However, D10 simplifies important variability that is associated with differences in particle ice nucleation effectiveness. For example, data used to derive D10 did not include marine aerosol measurements, and therefore D10 should not be expected to adequately predict INPs outside of regions dominated by continental aerosol, for example, in remote marine regions. This limitation can be overcome by the use of aerosol type-dependent parameterizations. Recent studies such as Shi and Liu (2019) and Fan et al. (2017) have used the parameterization of DeMott et al. (2015) to treat ice nucleation by mineral dusts. And increasingly aerosol-type-dependent deterministic parameterizations have been used to simulate $n_{INP}(T)$ arising from multiple aerosol sources (McCluskey et al., 2019; Penner et al., 2018; Phillips et al., 2008, 2013; Storelvmo et al., 2011).

3.1.2. Practical Challenges in Implementing INP Parameterizations in Models

As discussed in Section 2, when representing INPs in models, it would ideally be preferable to use a consistent set of parameterizations that treats all important particle types and spans the entire relevant range of temperatures and humidities. In practice, however, parameterizations are often applicable only for specific temperature and humidity conditions (e.g., Hiranuma, Hoffmann, et al., 2014; Hiranuma, Paukert, et al., 2014; Kulkarni et al., 2014, 2015; Niemand et al., 2012; Wex et al., 2014), in part due to instrument limitations and in part due simply to practical constraints on the amount of data that can be collected. Consequently, modelers must choose whether to extrapolate or truncate INP parameterizations when ambient temperatures and humidities are outside the experimentally validated range. Additionally, in some cases models have adopted a combination of theoretically inconsistent parameterizations (e.g., CNT for deposition freezing in combination with deterministic parameterizations for immersion freezing) due to a lack of a readily available comprehensive scheme for all particle types.

In addition to these limitations of INP parameterizations, another factor modelers must weigh is that implementing aerosol-aware INP parameterizations into aerosol-cloud microphysics schemes requires complex treatments of modeled aerosol. Complex aerosol emissions, loss mechanisms, and aerosol chemistry are computationally expensive to simulate and increase a model's conceptual complexity and software maintenance costs. However, increases in model complexity would be required to fully represent the many atmospheric processes that control INP number concentration. For example, many models employ highly simplified representations of aerosol mixing state (e.g., Liu et al., 2012, 2016). This simplification can preclude a model from distinguishing whether INPs are coated with soluble components (e.g., dust particles coated with organic matter), or whether these components are present as an external mixture. Therefore, a more complete representation of the impacts of atmospheric aging on ice nucleation may require a more detailed representation of particle mixing state than is supported by many current large-scale models (Riemer et al., 2019). Similarly, many large-scale models do not distinguish between interstitial and cloud-borne aerosol particles, that is, they do not predict what fraction of the particles in the cloudy region of a particular grid box are immersed in cloud droplets. Without this information, it is challenging to accurately simulate the contributions of the different modes of ice nucleation (immersion, deposition, and contact freezing), which critically depend on the aerosol's activation state (Wang et al., 2014). This limitation introduces fundamental uncertainties into simulations of INP-cloud effects in many large-scale models. Overcoming this challenge will require models to either more explicitly represent aerosol activation state and its subgrid variability, or develop improved algorithms for parameterizing its effects. More realistic representation of vertical velocity in models, including its small-scale turbulent fluctuations, is also important to simulate the ice formation efficiency in mixed-phase clouds (Bühl et al., 2019; Morrison et al., 2012). Modelers must weigh the costs of increased model complexity against any potential of demonstrated gains in realism when prioritizing model development needs.

3.1.3. Representativeness of Parameterizations for Regional or Global Applications

Another challenge is the fact that INP parameterizations are frequently derived under controlled laboratory conditions and from samples that may be representative only for a particular region. In many cases, more work is needed to understand whether these parameterizations are representative globally and in the real environment.

For example, Boose, Welti, et al. (2016) found that the ice-nucleating activity was considerably lower in airborne samples of Saharan dust compared with surface samples of Saharan dust that were later aerosolized in the laboratory. One likely source of such discrepancies is that methods used for laboratory aerosolization are imperfect

mimics of natural aerosolization; atmospheric processing may also play a role. This presents a fundamental challenge to the realism of laboratory-based INP parameterizations as well as for observational closure studies. Recent parameterizations that are derived from ambient measurements of isolated aerosol types, including DeMott et al. (2015) and McCluskey, Ovadnevaite, et al. (2018), overcome this challenge by avoiding the need for artificial aerosol generation, and by observing aerosol in ambient air that has already been exposed to atmospheric and physical and chemical processing. However, a major limitation of field-derived INP parameterizations is that particle types cannot be easily isolated in ambient samples and the impacts of atmospheric processing and aging cannot be easily quantified. Thus, to establish their predictive validity, both laboratory- and field-derived INP parameterizations require testing against independently obtained measurements (e.g., DeMott et al., 2015). Additionally, clearly defined descriptions of the context in which the INP parameterization can confidently be applied are critical for the use of INP parameterizations in modeling studies.

3.2. Model Simulation of INP Sources and Atmospheric Concentrations

INPs can arise from many sources, each associated with their own uncertainties and challenges that also translate to uncertainties in their respective ice-nucleating properties. A key challenge is incorporating adequate representations of the emission fluxes of relevant particle types. This requires development and evaluation of emission parameterizations that are linked to variables present in large-scale models, either in the form of empirical data sets (e.g., descriptions of land surface properties) or as predicted variables (e.g., surface winds and temperature). Once emitted, aerosols and their INP efficiencies evolve with mixing, transport, and cloud processing, requiring a careful assessment of transport and loss mechanisms to resolve cloud-level INP populations and their subsequent impacts on clouds (Figure 7). Gaps in current understanding of INP sources and atmospheric concentrations largely arise from experimental design constraints on the available observations (Section 2). Here we discuss (a) major INP sources and their challenges (Section 3.2.1) and (b) a critical need for vertically resolved INP concentrations and types (Section 3.2.2).

3.2.1. Key INP Sources and Their Challenges

To improve representation of INPs in models, advances are needed that improve understanding of (a) the INP activity of different aerosol particles (as a function of their composition and other properties), (b) the atmospheric concentrations and variability of those particle types that contribute to INPs, (c) how best to parameterize INP efficiency (n_s or J_{het}), and (d) how to most appropriately parameterize the aerosol sources, concentrations, and atmospheric evolution in models. While these four distinct aspects are comparatively well-understood for some INP sources (e.g., mineral dust), there are other particle types where the current level of understanding is far lower and advances are needed at a more fundamental level to improve current knowledge. While prior review articles focusing on INP particle types have largely focused on detailed discussions of the INP parameterizations and atmospheric measurements for each particle class (c above; Hoose & Möhler, 2012; Kanji et al., 2017; Murray et al., 2012), here we aim to provide a comprehensive evaluation that encompasses aerosol sources as well as their INP activities and does so from the perspective of both modeling and measurements.

Guided by current literature and our expert judgment, 10 aerosol types have been assessed for relative level of scientific understanding (RLOSU; Figure 8). We assess the RLOSU for four aspects of understanding: (a) observational constraints on aerosol abundance and composition; (b) modeling capabilities for aerosol emissions, processing, and composition; (c) measurement constraints on the aerosols' ice-nucleating ability; and (d) availability and representativeness of aerosol-specific INP parameterizations. The rubric that we have used to assign these RLOSU levels is provided in Supporting Information S1. In the remainder of this section, we briefly summarize and discuss the current status of scientific understanding, knowledge gaps, and research needs for each aerosol type. We also discuss the regions of the atmosphere, temperature regimes and ice nucleation modes, and the corresponding cloud types (mixed-phase or cirrus) for which each INP type is thought to be most relevant.

Desert dust is associated with particularly impressive ice-nucleating efficiency at temperatures cooler than −20°C (Kanji et al., 2017), and is arguably the best understood source of INPs across all four categories. Dust is a major source of variability in atmospheric INP concentrations and can be transported globally over long distances (Boose, Sierau, et al., 2016; Chou et al., 2011; DeMott, Sassen, et al., 2003). Mineral dust is a primary contributor to immersion-freezing ice nuclei in near-surface air, and is typically the dominant source at temperatures below

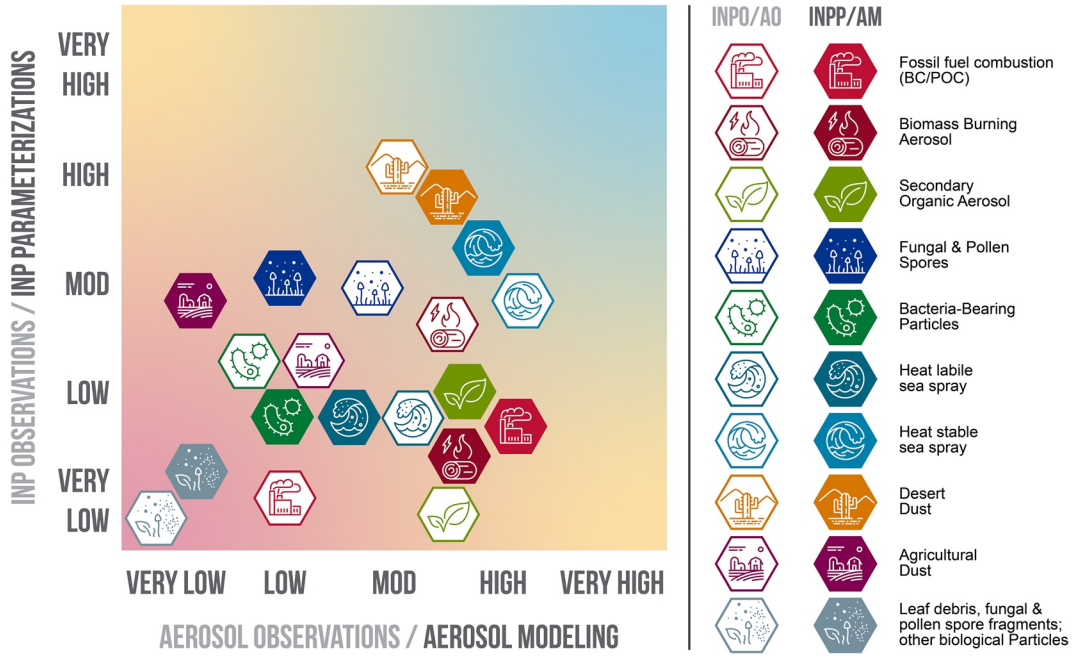

Figure 8. Summary of the relative level of scientific understanding based on available ice-nucleating particle (INP) observations, INP parameterizations, aerosol observations, and aerosol modeling for different particle sources contributing to atmospheric INPs. Each particle source is discussed in the text, with discussion of relevant literature in each area of aerosol and INP measurements and modeling that supports the selected ratings. Descriptions of the relative level of scientific understanding levels are provided in Supporting Information S1. Abbreviations are as follows: INPO, Ice-nucleating particle observations; AO, Aerosol Observations; INPP, Ice-nucleating particle parameterizations; AM, Aerosol Modeling.

−30°C (Murray et al., 2012). Mineral dust is also a dominant source of both immersion and deposition nuclei in the upper troposphere, and is an important contributor to cirrus cloud freezing nuclei (Cziczo et al., 2013).

Long-term measurements of dust mass concentration, size distribution, bulk composition, and optical properties are available from globally distributed in situ monitoring networks, alongside observations from numerous satellite-based and ground-based remote sensing instruments (Knippertz & Stuut, 2014; Rodriguez et al., 2012). Dust emission parameterizations have been incorporated into global climate models since the 1980s (Knippertz & Stuut, 2014). These parameterizations typically represent dust emissions as a function of soil properties (e.g., texture, particle size, threshold friction velocity for erosion), soil moisture, and surface wind speed (Albani et al., 2014; Ginoux et al., 2001; Kok, Albani, et al., 2014; Kok, Mahowald, et al., 2014; Zender et al., 2003). Increasingly, both soil data sets and dust representations in large-scale models have been evolving to include treatment of dust mineralogy (Claquin et al., 1999; Journet et al., 2014; Nickovic et al., 2012; Perlwitz et al., 2015a, 2015b; Scanza et al., 2015), which has been shown to play a role in determining the INP efficiencies of mineral dusts (Atkinson et al., 2013; Boose, Welti, et al., 2016). These model advancements have been met with dust-specific parameterizations, including both deterministic (DeMott et al., 2015; Niemand et al., 2012; Phillips et al., 2008) and stochastic parameterizations (Hoose & Möhler, 2012). The INP efficiency of mineral dust samples has been characterized in many laboratory studies, including a recent large measurement intercomparison, where illite-rich dust particles were characterized using 17 different measurement techniques (Hiranuma et al., 2015). Some recent INP parameterizations also begin to account for the effects of dust mineralogy on dust INP efficiency (Atkinson et al., 2013; Boose, Welti, et al., 2016) and progress has been made incorporating representations of dust mineralogy into surface dust data sets and global atmospheric models (Claquin et al., 1999; Journet et al., 2014; Nickovic et al., 2012; Scanza et al., 2015).

Despite this high level of understanding, gaps remain in our understanding of mineral dust aerosols and their activity as INPs, and in our capabilities to represent them in models. For example, Boose, Welti, et al. (2016) found differences of an order of magnitude between the INP activity of laboratory-generated dust samples and samples of ambient dust aerosol. From the experimental side, additional effort is needed to understand these

differences. Long-term monitoring of INP concentrations in parallel to characterization of mineral dust can also add value through closure studies that test the robustness of laboratory-derived and field-derived mineral dust INP parameterizations for ambient samples in locations impacted by the world's major dust sources.

Because dust is an effective source of INPs, we also highlight a need to further understand how dust impacts remote regions. Global models exhibit significant diversity in their ability to accurately simulate dust concentrations, especially in regions remote from dust sources (Huneeus et al., 2011). Recent studies have suggested that most current global models may overestimate dust removal (Ridley et al., 2016) and underpredict concentrations of supermicron dust particles (Adebiyi & Kok, 2020; Kok, 2011). Continued effort is needed to evaluate and improve dust aerosol physics, including size distribution, loss mechanisms, and optical properties, and application of dust INP parameterizations to aged and transported dust aerosols in large-scale models (Kok, Albani, et al., 2014).

Agricultural aerosol and organic-rich soil dusts. Organic-rich soils from arable lands have attracted increasing attention in laboratory and field experiments as evidence for their potential atmospheric importance. Multiple laboratory studies have found that organic-rich soils are more INP active than inorganic desert dusts in both the immersion and deposition modes (Conen et al., 2011; O'Sullivan et al., 2014; Steinke et al., 2016; Suski, Hill, et al., 2018; Tobo et al., 2014). This raises the possibility that soils aerosolized by agricultural operations may largely determine local INP concentrations in agricultural regions, where agricultural dusts may be the dominant source of dust emissions and dust aerosol optical depth (Chen et al., 2018; Ginoux et al., 2012). Although some emissions inventories targeted towards air quality modeling include estimates of agricultural dust (e.g., the United States Environmental Protection Agency's National Emissions Inventory; Penfold et al., 2011; Tooly, 2001), agricultural dust emissions are not yet routinely represented in any climate or Earth system model to the best of our knowledge, since these models typically only account for natural wind-blown dust emissions from arid regions (Kok, Mahowald, et al., 2014; Mahowald, 2011; Zender et al., 2003). Additionally, dust emissions in future climate projections are subject to feedbacks from the effects of climate change as well as uncertainties in response to future shifts in human land use, agricultural management, and water usage (Muhs et al., 2014; Tegen & Fung, 1995; Tegen et al., 2004; Webb & Pierre, 2018; Woodward et al., 2005). Intriguing new evidence suggests that the organic-rich soil of the thawing Arctic permafrost is a remarkably strong source of INPs, suggesting a novel and previously overlooked pathway by which future changes in INP sources may impact clouds and climate (Creamean et al., 2020).

Given the impressive ice nucleation behavior of agricultural aerosol organic-rich soils, further studies are needed that quantify their atmospheric concentrations and test our ability to predict their contribution to atmospheric INP concentrations through closure experiments (Knopf et al., 2021). Additionally, more work is needed to link INP properties of agricultural dusts to the detailed particle composition, since particles with different sizes and different organic and biological components may potentially nucleate ice differently. Complementary studies that connect the detailed composition of individual particles to INP efficiency can enable parameterizations that quantify this relationship, which in concert with agricultural soil aerosol emission parameterization development, could pave the way for improved representation of these INPs in future model development efforts.

Primary biological aerosol particles (PBAP) include whole or fragments of cellular material that is aerosolized in the environment, such as pollen, bacteria, fungal spores, and plant debris (Burrows, Elbert, et al., 2009; Després et al., 2012; Elbert et al., 2007). PBAP can be emitted passively—through wind-driven emissions or by being dislodged from leaf surfaces following raindrop impacts—or emitted actively by ejection from certain fungi. Such particles are present in low concentrations in the atmosphere, but their role as INPs has been a topic of significant research interest due to the high freezing efficiency of some biological particles at warmer temperatures (Iannone et al., 2011; Morris et al., 2004). The contribution of biological INPs is often inferred indirectly by quantifying the loss of INP activity in samples that are exposed to heat (which can denature complex biological macromolecules), to enzymes such as lysozymes (which breaks down bacterial cell walls), and other treatments (Hill et al., 2016). Field experiments have found correlations between measurements of biological particles and collocated measurements of INPs; in some cases, observed peaks in both PBAP and INPs have been tied to rainfall events (Huffman et al., 2013; Mason et al., 2015; Tobo et al., 2013). Biological particles may be an important source of INPs in regions where they are plentiful, such as the Amazon (Prenni, Petters, et al., 2009). Additionally, the broader term biogenic aerosol is sometimes used, in order to be inclusive of organic aerosol types of biogenic origin that do not fit within the definition of PBAP (e.g., exopolysaccharides and biogenic secondary

organic aerosol or SOA). Recently, long-term measurements of INPs in the Finnish boreal forest identified a pronounced seasonal cycle in INP concentrations, with higher concentrations during summer, which they attributed to biogenic particles (Schneider et al., 2021).

INP parameterizations are available for specific types of biological particles, including deterministic (Patade et al., 2021; Tobo et al., 2013) and CNT-based (Hoose, Kristjánsson, Chen, & Hazra, 2010). Laboratory experiments have characterized biological particles in the contact freezing (Niehaus et al., 2014) and deposition nucleation modes (Hoose & Möhler, 2012). However, the focus of most studies of PBAP as INPs has focused on immersion freezing (Hoose & Möhler, 2012); this is assumed to be the mode in which they are most atmospherically relevant, due to their particularly strong freezing efficiency at the warmer temperatures characteristics of mixed-phase clouds (Hoose, Kristjánsson, & Burrows, 2010). These parameterizations, while useful for modeling studies, should be regarded as having significant caveats. In particular, the INP activity of biological particles can vary dramatically both between and within taxa, and in response to environmental conditions (Burrows, Elbert, et al., 2009; Després et al., 2012). Many of the studies that parameterized the INP efficacy of biological INPs have used a "bottom-up" approach, by developing parameterizations for individual taxa. Because of the large variability between taxa, the large number of taxa present in the atmosphere and the uncertainty surrounding their behavior under atmospheric conditions, the bottom-up approach faces practical challenges when aiming to develop atmospherically representative parameterizations for biological INPs that can be applied in models. To overcome these challenges, some recent efforts have begun to take a "top-down" approach, by inferring the INP efficiency of biological particles directly from atmospherically collected samples (Patade et al., 2021). Applications of similar approaches across diverse ecosystems, together with closure studies to test whether these parameterizations can improve INP predictability, could prove to be fruitful future research directions.

Biological particle fragments: Recently, laboratory experiments have also shown that small fragments of biological particles (e.g., bacteria, fungal spores, and pollen) can serve as INPs (Augustin et al., 2013; Pummer et al., 2012, 2015; Suski, Bell, et al., 2018). Leaf litter has also been shown to be a potent source of INPs (Schnell & Vali, 1976). However, quantifying their prevalence in the atmosphere is experimentally challenging. Fungal and pollen fragments can be produced when spores and pollen burst under high humidity conditions (China et al., 2016; Lawler et al., 2020). An early modeling study that aimed to simulate pollen-bursting events suggests that during spring pollen season in the United States, concentrations of sub-pollen particles can range from approximately 1 to 1,000 cm^{-3}, potentially suppressing seasonal precipitation through their activity as CCN, depending on the number of particles produced per pollen grain (Wozniak & Steiner, 2017; Wozniak et al., 2018). Concentrations of this magnitude could potentially contribute significantly to INP concentrations on a seasonal and regional basis. However, few if any in situ measurements have been made that quantify fungal or pollen fragment abundance in the atmosphere, which would be needed to evaluate the realism of such modeling results. Quantitative measurements of the atmospheric abundance of nanometer-scale atmospheric fungal and pollen fragment is an experimental challenge, but would provide a useful constraint on the potential for these fragments to contribute to atmospheric freezing processes.

Incorporation of bacteria, fungal spore, and pollen INPs into models: Global emissions parameterizations have been developed for bacteria-bearing particles (Burrows, Butler, et al., 2009), fungal spores (Heald & Spracklen, 2009; Janssen et al., 2021), and pollen (Wozniak & Steiner, 2017). In the case of pollen, there are also a number of regional-scale emission parameterizations, but these tend to be focused on applications to human allergen forecasting and so simulate only a subset of all pollen species (e.g., Helbig et al., 2004; Prank et al., 2013, 2016; Scheifinger et al., 2013; Sofiev et al., 2015; Zink et al., 2012). There are relatively few quantitative measurements of biological particles worldwide, with almost no observations available in some regions of the world (Burrows, Elbert, et al., 2009; Buters et al., 2018). This relative lack of observational data, in combination with inherent complexity of modeling ecosystem processes, should temper expectations for the accuracy and precision of simulations of biological particle concentrations. Parameterizations of biological particle emissions have been evaluated against independently collected observational data sets with mixed results. Models may generally be able to simulate mean number and mass concentrations of biological particles within the correct order of magnitude (e.g., Burrows, Rayner, et al., 2013; Janssen et al., 2021). In some cases they have been shown to simulate seasonal cycles or regional-scale geographic distributions that correlate with observed patterns, but tend to lack skill in reproducing day-to-day variability (Perring et al., 2015; Twohy et al., 2016). For this reason, the currently available parameterizations are likely to be more informative for gaining an order of magnitude understanding

of their potential relative contributions to atmospheric INP in different regions, and potential cloud impacts in a climatological sense, than for applications that require greater skill in capturing temporal and geographic trends such as short-term weather forecasting. As the community gains an improved understanding of the relative role and importance of biological INPs in the climate system, it is conceivable that additional advances in these emission parameterizations may become useful. However, in the near term, process-level studies that use novel approaches to collect and analyze field campaign data quantifying biological particle contributions to INPs, and which can be used to evaluate and more clearly identify shortcomings of the existing parameterizations, may prove more fruitful in advancing understanding of the atmospheric relevance of these INPs.

Early studies incorporating biological INPs into global models to study their impact on cloud processes have indicated that biological particles may contribute significantly to initiating freezing in the lowest (and warmest) portions of mid-latitude clouds, but have negligible impact on freezing rates in the higher altitude, colder portions of clouds (Hoose, Kristjánsson, & Burrows, 2010; Spracklen & Heald, 2014). However, the importance of this early freezing for the overall cloud glaciation process is not yet fully understood. Cloud-resolving models have suggested that mid-latitude, mixed-phase clouds may be sensitive to biological INPs when they are present in concentrations significantly exceeding their typical background concentrations (Phillips et al., 2009). However, the sensitivity of clouds to warm-temperature INPs likely depends significantly on the strength of secondary ice production (SIP) processes, discussed further in Section 3.4. Therefore, studies of SIP are needed that employ cloud-resolving models, and in-cloud measurement of INPs and hydrometeors, to improve understanding of the role of SIP at a fundamental level.

Biomass-burning aerosol, comprising black carbon, organic aerosol, mineral dust, and plant debris, has been identified as a complex INP source (e.g., Petters et al., 2009). Measurements of biomass burning aerosol are available from ground-based observation networks (e.g., IMPROVE/AIRNOW and AERONET), airborne (e.g., Guyon et al., 2005), and surface observations. The ice-nucleating activity of biomass-burning aerosol is highly variable and often significantly lower than that of mineral dust (by two orders of magnitude or more), but has been a subject of research interest due to the high concentrations of aerosol that can be present in biomass-burning plumes. By applying field measurement constraints on the efficacy of biomass burning INPs with simulations of atmospheric biomass burning aerosol concentrations (Schill et al., 2020), similarly concluded that biomass burning black carbon particles contributed minimally (about 5%) to INP populations on a global average. Therefore, the impacts of biomass burning INPs are expected to be localized and episodic, with comparatively limited relevance at global scales.

Extensive laboratory experiments have been performed to characterize the INP activity of biomass-burning aerosol produced from a variety of source plant materials (McCluskey et al., 2014; Petters et al., 2009) and biomass-burning INPs have been characterized in the field (Barry et al., 2021; McCluskey et al., 2014). However, it is difficult to attribute INP activity in ambient biomass-burning particles to specific aerosol types, since in these plumes a variety of types of particles (e.g., soot, organic particles, dust) are often commingled (McCluskey et al., 2014). Additionally, field and laboratory studies have shown that biomass burning plumes undergo complex chemical transformations during transport that models cannot yet capture, which may result in modified ice-nucleating activity (Hodshire et al., 2019; McMeeking et al., 2009). For example, Schill et al. (2020) and Barry et al. (2021) show that organic components are a potentially useful predictor of INPs active at temperatures warmer than −25°C in biomass-burning plumes, which may be associated with in situ production of tarballs. Another recent study by Jahl et al. (2021) finds surprising evidence that the ice-nucleation ability of biomass-burning aerosol may be enhanced during atmospheric transport and aging.

Many regional and Earth system models represent biomass-burning aerosol; for example, the modal aerosol model (as implemented in the CESM2, WACCM, and E3SM GCMs) includes emissions of black carbon, SOA, and particulate organic carbon from biomass burning. However, a recent detailed model evaluation exercise by Reddington et al. (2019) concluded that additional, detailed observations of aerosol properties over biomass-burning regions are required to improve constraints on the satellite-derived fire emissions data sets typically used in such models, such as the Global Fire Emissions Database (GFED; Randerson et al., 2015), Global Fire Assimilation System (GFAS; Kaiser et al., 2012), Quick Fire Emissions Data set (QFED; Darmenov & da Silva, 2015), and Fire INventory from NCAR (FINN; Wiedinmyer et al., 2011) inventories. Additionally, these aerosols, although present in models, are not necessarily linked to INP parameterizations and cloud ice formation.

Additional research will be needed to quantify the relative importance of this complex chemistry for INP impacts on cloud freezing over both continental and remote regions.

The IN activity of *sea spray aerosol* is two or more orders of magnitude lower than that of mineral dust (DeMott et al., 2016), and is associated with the organic components of the sea spray particles. Historically, sea spray was largely considered unimportant as a source of atmospheric INPs; however, during the past decade, sea spray aerosol has been clearly established as an important INP source in marine regions remote from continental aerosol sources (Burrows, Hoose, et al., 2013; McCluskey et al., 2019; McCluskey, Hill, Humphries, et al., 2018; Vergara-Temprado et al., 2017; Wilson et al., 2015). Recent studies have experimentally confirmed the existence of marine organic ice-nucleating material in natural seawater and sea surface microlayers (Irish et al., 2019; Wilson et al., 2015). Studies of laboratory-generated sea spray aerosol have identified multiple particle types that contribute to marine organic INP, including (a) dissolved organic carbon INPs that are suggested to be composed of heat-stable ice-nucleating active molecules (e.g., long-chain fatty acids DeMott, Mason, et al., 2018) and are omnipresent ("heat stable sea spray" INPs in Figure 8) and (b) particulate organic carbon INPs, including heat-labile intact cells (e.g., Knopf et al., 2011) or ice-nucleating active microbe fragments, which are episodically present (McCluskey, Hill, Sultana, et al., 2018, "heat labile sea spray" INPs in Figure 8). Field measurements have also provided evidence for these marine organic INPs in marine air masses, likely associated with local and regional ocean biological activity (Creamean et al., 2019; McCluskey, Ovadnevaite, et al., 2018). While most studies of sea spray INPs have focused on the immersion mode due to the relevance of sea spray to marine mixed-phase clouds, aerosol particles generated from seawater samples have also been shown to nucleate ice in the deposition mode (Wolf, Goodell, et al., 2020).

An early parameterization of sea spray INP based on material collected from the sea surface microlayer was developed by Wilson et al. (2015) to predict INPs based on total organic carbon and ice-nucleating temperature. The sea-air transfer of sea spray organic INPs is a major source of uncertainty when applying the Wilson et al. (2015) parameterization to ambient or simulated sea spray aerosol. Enrichment of organic and biological materials in sea spray is both considerable and highly variable (Burrows et al., 2014). Although the majority of ice-nucleating entities in bulk and surface seawater samples are between 0.02 and 0.2 μm diameter (Irish et al., 2017; Wilson et al., 2015), an intriguing recent study finds that supermicron sea spray particles have higher n_s than submicron sea spray particles (Mitts et al., 2021). Large uncertainties persist regarding both the efficiency with which ice-nucleating entities are transferred to supermicron sea spray, and the efficacy of larger sea spray particles as INPs, currently limiting our ability to confidently determine whether supermicron sea spray INPs are important players at regional and global scales (Steinke et al., 2021). A parameterization of sea spray INP activity based on direct measurements of ambient sea spray was developed by McCluskey, Ovadnevaite, et al. (2018) to predict the background marine heat-stable dissolved organic carbon INP type based on total sea spray aerosol surface area and ice-nucleating temperature. McCluskey et al. (2019) demonstrated potential for global aerosol models to adequately predict concentrations of INP in the remote marine boundary (e.g., the Southern Ocean) when using this parameterization.

Regional and global atmospheric models have long represented sea salt aerosol (i.e., the inorganic components of sea spray), but have only recently begun to include parameterizations of the organic components of sea spray that are associated with sea spray aerosol's ice-nucleation activity, for example, Vignati et al. (2010), Long et al. (2011), Gantt et al. (2011), and Burrows et al. (2014). Most recently, modeling studies have applied existing marine organic aerosol INP parameterizations (McCluskey, Hill, Sultana, et al., 2018; Wilson et al., 2015) to modeled sea spray aerosol (McCluskey, Ovadnevaite, et al., 2018) and evaluated their impact on simulated clouds as CCN and INPs (Burrows, Easter et al., 2022, Vergara-Temprado et al., 2018; Huang et al., 2018; X. Zhao et al., 2021).

Through this past decade of focused community research effort, a clear understanding has emerged that sea spray aerosol is an important background source of INPs in marine boundary layer air in regions that are remote from continental aerosol sources, such as the Southern Ocean. However, knowledge gaps remain in understanding the seasonal cycles of sea spray aerosol INP abundance, the impacts of ocean biological activity on the ice-nucleating efficiency of sea spray, and the extent to which sea spray aerosol is relevant at cloud altitudes, where models suggest that long-range dust transport may also contribute to the INP population (McCluskey et al., 2019; Vergara-Temprado et al., 2017). However, as discussed previously, simulated dust concentrations in remote regions are highly uncertain.

Other recent efforts have focused on anthropogenic pollution aerosol as a potential source of INPs. Anthropogenic pollution aerosols can include a mixture of SOA, soot, lead and other metal oxides, and solid sulfate.

Limited but intriguing early results from laboratory studies have suggested that SOA particles, which may originate from either anthropogenic or biogenic precursor gases, may serve as deposition INPs (Wang, Lambe, et al., 2012), when they become "glassy" under the cold, dry conditions typical of the upper troposphere (Knopf et al., 2018; Shiraiwa et al., 2017). Wolf, Zhang, et al. (2020) recently showed evidence of isoprene-derived SOA contributing to atmospheric depositional INP concentrations at cirrus temperatures during measurements at a mountaintop site. Very low active fractions of such particles were confirmed in related laboratory measurements. Hence, although this research could have significant implications for cirrus when SOA is present in high number concentrations at cirrus levels, SOA involvement in cirrus formation has yet to be confirmed through in situ observations.

Fossil fuel combustion aerosols, including soot and other black carbon aerosols, as well as particulate organic carbon aerosols, have also been discussed as a potential source of anthropogenic INPs. For the mixed-phase cloud regime, recent laboratory evidence indicates that immersion-mode freezing by soot particles is weak and that soot therefore can be neglected for ice formation in the atmosphere at temperatures warmer than −38°C (Kanji et al., 2020).

Fossil fuel combustion aerosol is thought to be more relevant to the cirrus cloud regime, where soot can contribute to deposition freezing nuclei, and where combustion aerosol from aircraft emissions is a significant source of particulate matter in otherwise pristine air. Cirrus cloud microphysical behavior is governed by competition between the rates of homogeneous freezing and heterogeneous freezing facilitated by INPs (discussed further in Section 3.4). Because direct measurements of cloud ice residuals in the upper troposphere are challenging, however, the argument for the importance of both SOA and soot to cirrus ice formation globally thus far has been made largely on the basis of models, using results from laboratory experiments to parameterize particle freezing efficiency. In a recent example, Zhu and Penner (2020) examined the effects of aircraft soot, sulfur emissions, and other anthropogenic emissions on cirrus clouds in a variant of the CESM model.

The strength of effects predicted in such modeling studies depends critically on the assumptions used regarding the freezing efficiency of INPs. Most recent studies agree that freshly formed soot particles are only weakly active as INPs in the deposition freezing regime (Dymarska et al., 2006). The freezing efficiency of soot is sensitive to atmospheric aging processes, including cloud processing, water uptake, and coating with condensable vapors (Friedman et al., 2011; Kulkarni et al., 2016; Möhler et al., 2005), but these processes in general do not appear to significantly increase the efficacy of soot as a freezing nucleus. However, a recent study by Mahrt et al. (2020) found that the efficiency of soot particles acting as deposition freezing nuclei at $T < 233$ K was greatly enhanced after pre-activation by cloud processing, potentially increasing the relevance of these particles for freezing in cirrus.

Ultimately, a major outstanding challenge in constraining the effects of soot, SOA INPs, and other anthropogenic aerosol components on cirrus cloud behavior is the paucity of observational constraints from atmospheric observations. Direct aircraft-based measurements of cloud properties and cloud ice residuals in the upper troposphere are required to constrain models, and more firmly establish what role anthropogenic SOA or soot INPs may play in impacting Earth's radiative budget.

3.2.2. A Critical Need for Vertically Resolved INP Concentrations and Types

Most measurements used to constrain numerical INP simulations have occurred near the Earth's surface, with far fewer observational constraints available for the concentrations of INPs at cloud altitudes. In simulating aerosol and INP concentrations in clouds, models rely on their own simulation of particle vertical transport and removal processes. Models of atmospheric INPs can also be improved by studies that provide observational constraints on the atmospheric processes that control their transport and removal. For example, recent measurements of the vertical profile and long-distance transport of supermicron Saharan dust particles revealed significant biases in model transport of these particles (Adebiyi & Kok, 2020), the causes of which have yet to be untangled.

Vertical tracer transport in models occurs both through resolved processes (e.g., frontal uplift in synoptic-scale weather systems) and unresolved subgrid processes, which must be parameterized (i.e., boundary layer turbulence, gravity waves, and in most present-day global model simulations, moist convection). These parameterized

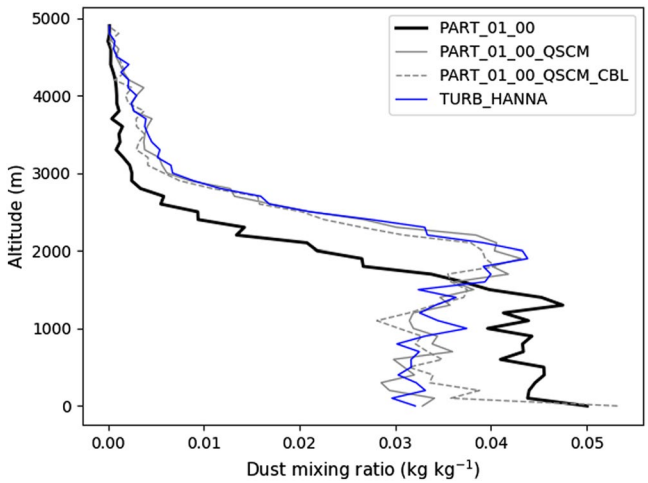

Figure 9. Vertical profiles of mixing ratios for a surface-emitted 1 μm diameter dust tracer at the end of a day with simulated shallow convection. The base case simulation (PART_01_00), with 100 m grid spacing, is compared with a quasisingle column model (QSCM) simulation (PART_01_00_QSCM), a QSCM with the convective boundary layer (CBL) namelist option turned on (PART_01_00_QSCM_CBL), and with the Hanna turbulence parameterization (PART_01_00_HANNA). The difference between the vertical profiles for the base case simulation where CBL turbulence is resolved by the underlying LES simulation, as compared with the other cases where turbulence is not resolved, is illustrative of the critical role of turbulence parameterizations in controlling particle vertical transport, and the potential uncertainties that these parameterizations can introduce in the simulation of INP transport into clouds. Reproduced from Cornwell, Xiao, et al. (2021).

processes introduce significant uncertainties in simulated high-altitude particle concentrations. In particular, vertical transport associated with boundary layer turbulence is one of the most important factors explaining intermodel differences in aerosol vertical profiles, even at the highest altitudes Kipling et al. (2016). The efficiency with which aerosol particles are exported from the boundary layer can differ significantly between LES simulations, which resolve most boundary layer turbulent eddies, and simulations with parameterized turbulence. For example, Cornwell, Xiao, et al. (2021) simulated vertical transport of a Lagrangian particle ensemble using meteorological output from an LES simulation (grid spacing: 10 km), and found significant differences in the vertical export of particles, as compared to a simulation of the same day that mimics a single column of a regional-scale model by averaging meteorological conditions across the entire 25 km domain and applying a turbulence parameterization (Figure 9). Parameterized deep convective mass transport can also have important impacts on the simulated vertical and long-range transport of atmospheric trace constituents, including INPs (Tost et al., 2010).

More constraints may be provided by the growing trend toward vertical profile measurements of aerosol and INP using tethered balloons (Creamean, de Boer, et al., 2020; Lawson et al., 2011; Porter et al., 2020) and unmanned aerial vehicles (Bieber et al., 2020; Marinou et al., 2019; Schrod et al., 2017). These technologies enable a longer sampling duration than aircraft-based measurements, and so can offer a better platform for capturing statistically significant sample sizes of INPs.

Remote sensing measurements have increasingly been exploited to understand INP vertical distributions and their atmospheric impacts. Ground-based and space-based lidar sensors are particularly advantageous for such studies since they rely on the reflectance of an actively pulsed laser to measure both particle and cloud properties. This active sensing method can retrieve more independent pieces of vertical information than passive sensing techniques. For example, in lidar measurements of supercooled altocumulus clouds in Alaska, Sassen and Khvorostyanov (2008) found evidence of glaciation induced by entrainment of smoke aerosol from boreal forest fires. In another example, Tan et al. (2014) used space-borne lidar measurements to examine relationships between the vertical profiles of cloud phase, dust, polluted dust, and smoke aerosols. They found that while temperature was the dominant factor controlling cloud phase, dust and potentially smoke aerosol were also correlated with decreases in supercooled liquid cloud fraction.

Using observations from multiple satellites in combination with cloud-resolving modeling, Zhao et al. (2019) developed a new method to evaluate the ice nucleation ability of aerosols of different origins by examining the bulk relationships between aerosol loading and ice crystal size. Their results suggest that polluted continental aerosols in Asia contain a fraction of INPs, though the exact chemical compositions of these INPs are yet to be explored. Finally, Ansmann and colleagues have used polarization lidar to derive profiles of aerosol type, concentration, and surface area, and combined these with existing parameterizations to produce vertical profiles of INP from desert dust, marine, and continental dust INP types (Ansmann et al., 2019; Mamouri & Ansmann, 2015, 2016). In another study, elevated INP concentrations measured on board an unmanned aircraft system coincided with detection of dust plume by lidar backscattering, building further confidence in the utility of lidar for remote sensing of dust INPs (Schrod et al., 2017).

Aerosol loss mechanisms create additional complications for the simulation of atmospheric INP concentrations. Parameterized loss processes include dry deposition to the Earth's surface (e.g., Farmer et al., 2021) as well as wet removal due to both activation scavenging and impaction/interception scavenging (e.g., Croft et al., 2009; Grythe et al., 2017; Tost et al., 2007; Yang et al., 2015). Errors in loss processes—including those caused by structural aspects of models such as inaccurate representation of the particle size distribution or model grid resolution—can significantly impact simulated particle atmospheric residence times (Burrows, Rayner, et al., 2013; Croft et al., 2012, 2014; Emerson et al., 2020; Saylor et al., 2019; H. Wang et al., 2013; X. Wang et al., 2010)

and change the proportion of simulated particle mass that is present distant from sources, as compared with near-source concentrations (Abdelkader et al., 2017; Haga et al., 2014; H. Wang et al., 2013; Wu et al., 2020).

Dry deposition can be an important source of uncertainty for predicting airborne and deposited particulate matter, contributing both to parametric uncertainty within individual models (Carslaw et al., 2013; Fanourgakis et al., 2019), and to intermodel differences (Im et al., 2015). Dry deposition of particles in most state-of-the-art models is computed as a function of particle size using the theoretical "resistance" framework established by Slinn (1982). In this approach, the processes controlling the rate of mass deposition to the surface are treated analogously to sources of resistance in an electrical circuit that operate either in serial or in parallel. The deposition velocity, v_d, is typically calculated as:

$$v_d = v_s + \frac{1}{(r_a + r_s)} \tag{1}$$

where v_s is the particle's gravitational settling velocity, r_a is the aerodynamic resistance, and r_s is the resistance of the surface (or canopy).

However, despite using a common conceptual framework, models use different approaches to parameterize the individual resistance terms, leading to substantial discrepancies in predicted deposition velocities (Saylor et al., 2019). Modeling of dry deposition is also complicated by challenges in generalizing from field measurements; for example, measurements made at one location may not be representative for other locations (Hicks, 1995) and complex terrains can increase dry deposition velocities (Hicks, 2008). Since larger particles are more likely to both act as INPs and be lost through dry deposition, uncertainties in the treatment of dry deposition could impair the accurate simulation of INPs. Indeed, recent work suggests that widely used model parameterizations may significantly overestimate dry deposition of accumulation-mode particles and underestimate dry deposition of coarse-mode particles (Emerson et al., 2020). For a recent review of dry deposition mechanisms, measurements, and field observations, see Farmer et al. (2021).

Accurate simulation of wet removal is challenging due to the inherently multiscale nature of aerosol-cloud interactions, which can introduce large uncertainties into simulated aerosol concentrations and their global distribution. For example, one study estimated that the parameterization of wet removal of entrained aerosol in deep convective systems introduced an error of up to 35% in simulated global, annual mean aerosol optical depth (Croft et al., 2012). Additionally, coarse-resolution models are unable to represent the sub-grid filamentary patches of clear air within precipitating cloud systems, which can harbor aerosol particles that are not removed (Ma et al., 2013). Typically, the values of uncertain parameters in global models are calibrated in each new model release in an effort to obtain reasonable agreement with large-scale, climatological observations such as satellite-observed aerosol optical depth, which can potentially result in errors in the relative concentrations of near-source and remote aerosol (e.g., Wu et al., 2020).

In addition to the challenges inherent to simulating removal of all aerosol species, there is a special challenge particular to simulation of INPs: representing the preferential removal and eventual depletion of INPs in mixed-phase clouds. The impacts of preferential scavenging can be immediate or could occur downstream by modifying the particle atmospheric residence times. Cloud-resolving model studies of long-lived Arctic mixed-phase boundary layer clouds have shown that preferential scavenging can lead to extremely low concentrations of INPs in these clouds. Simulating this depletion has proven critical for obtaining credible simulations of these clouds in cloud-resolving models (Avramov & Harrington, 2010; Fan et al., 2009; Fridlind et al., 2007; Harrington & Olsson, 2001; Morrison et al., 2005; Prenni, Harrington, et al., 2007). Preferential scavenging of INPs may also significantly reduce their concentrations during long-range transport (i.e., distant from sources), especially impacting concentrations in the upper troposphere and at high latitudes (Haga et al., 2014; Jensen et al., 2018).

For global climate models, state-of-the-art, aerosol-cloud microphysics schemes often explicitly represent the fraction of aerosol that is immersed in liquid cloud droplets but most do not explicitly represent the ice-borne aerosol. Consequently, many models do not include any mechanism to simulate preferential scavenging of INPs, a capability sometimes referred to as "prognostic INPs" (see Section 1.1.9 in the supplement of Liu et al. [2012]). This can potentially have large impacts on the atmospheric residence time, vertical distribution, and long-distance transport of these particles (Haga et al., 2014).

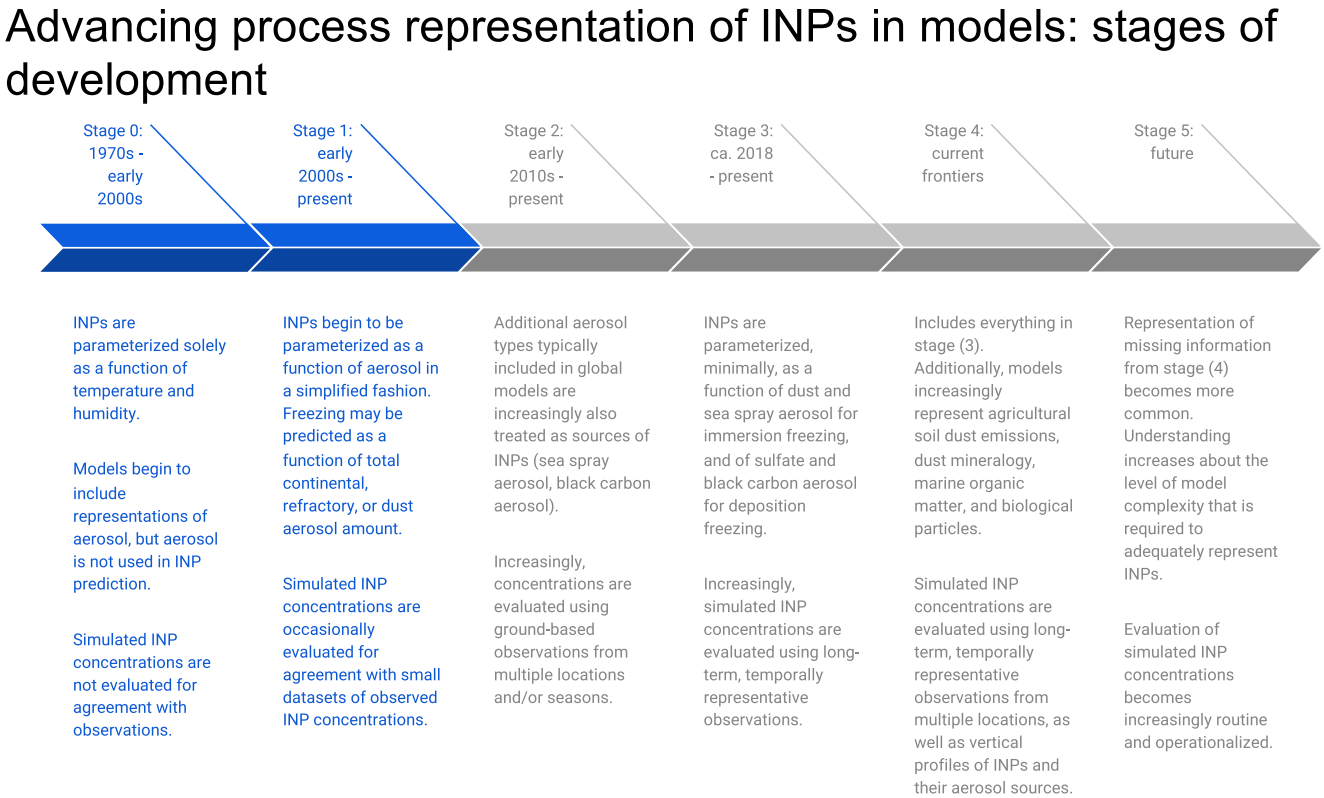

Figure 10. Stages of development toward improving realism of ice-nucleating particle (INP) representations in atmospheric models and approximate years in which each stage begins. Note that because models have chosen different tradeoffs in terms of the level of complexity they represent in aerosol and cloud microphysical processes, present-day models exist at all stages (0–5), with models that prioritize simplicity represented in Stages 0 and 1, and models that emphasize greater realism and complexity in aerosol and cloud ice microphysics represented in Stages 2–4. In Stage 5, we envision the needs of future modeling efforts that aim to build and evaluate increasingly robust INP representations.

Errors in these processes disproportionately impact simulated INP concentrations in remote high-latitude regions and at high altitudes; these are often the same regions that are characterized by a relative scarcity of INPs, such that INP availability may be rate-limiting for ice formation in the same clouds most impacted by the representation of INP preferential scavenging processes. Several recent field campaigns have observed aerosol properties and characteristics at high latitudes (e.g., the Antarctic Circumpolar expedition, SOCRATES, MICRE, MARCUS, AWARE, and MOSAIC; see McFarquhar et al. [2021], for an overview of several of these campaigns). However, relatively few concurrent measurements are available of aerosol and INP vertical profiles. Therefore, there is a particularly critical need for measurements of the vertical profiles of INPs and their source particles. Additional observational needs for the evaluation and validation of INPs in models are discussed in the following section.

3.3. Constraining Simulations of INP Number and Evaluating Model Skill

A critical step in implementing INP parameterizations into models is the evaluation of the model's skill in simulating INP concentrations that agree with those measured in the ambient atmosphere. “Skill” is defined by the Glossary of Meteorology as “A statistical evaluation of the accuracy of forecasts” (Glickman & Zenk, 2000), and can be quantified by various scalar metrics, such as those recommended by Huijnen and Eskes (2012) for forecasts of atmospheric composition.

As aerosol-aware INP parameterizations are increasingly implemented into models, there is a growing need for observational data sets that can be used to appropriately evaluate such parameterizations. Increasingly, models require aerosol-INP evaluation data sets that capture greater process detail and are more geographically and temporally representative. In the future, evaluation of INPs and their major particle sources could be increasingly operationalized, for example, through the increased development and use of community benchmarking data sets and tools (Figure 10).

Ideally, every study that implements a newly developed INP parameterization in a regional or global model would demonstrate that the new representation produces an objective improvement in the model's skill in forecasting INP number concentrations. In practice, our experience with utilizing published field measurements to constrain and evaluate global models of INPs reveals several challenges that may make such efforts prohibitively laborious. To address this gap, we offer recommendations for the INP measurement community to consider when designing and publishing data from field campaigns that are intended for use in model evaluation.

A first-order consideration is the timing and location of the INP measurements. Perhaps the most important obstacle to the use of INP measurements for large-scale model evaluation is their limited spatial and temporal representativeness. In comparison with CCN and air quality variables, INPs and the subset of aerosol particles that contribute to their atmospheric concentrations have been measured far less frequently, in fewer locations, and for shorter time periods (typically days to weeks). Considering the high temporal variability of atmospheric aerosol concentrations, it is unlikely that typical field campaigns, lasting 1–4 weeks, provide an adequate sampling of INP temporal variability to fully evaluate model skill. Long-term field measurements of INPs have only recently begun to be conducted (e.g., Lacher et al., 2018; Schneider et al., 2021; Schrod et al., 2020); these studies are revealing new insights into both the short-term and seasonal variability of INP concentrations. More long-term measurements should be conducted at carefully selected sites that can provide information on a variety of INP types and sources; such measurements are likely to be facilitated by new and emerging measurement technologies developed during recent years (Brunner & Kanji, 2021; Möhler et al., 2021; Tarn et al., 2020). Remote sites may be particularly helpful in understanding the INP variability in regions less impacted by anthropogenic aerosol sources (Schrod et al., 2020) and alpine sites that can sample free tropospheric air may be helpful in understanding the concentrations of INPs at higher altitudes (Lacher et al., 2018).

The heterogeneity of published INP measurements with respect to measurement methods and metadata also inhibits their use for evaluation of model skill. INP measurements can provide significantly greater scientific insight when made available in combination with concurrent aerosol measurements, ideally including measurements of size-resolved and particle-resolved composition as discussed in Section 2.3. These measurements are critical to discriminate between errors in simulation of aerosol amount and errors in the parameterized ice-nucleating efficiency of the aerosol. Additionally, experiments that include direct chemical characterization of INPs provide a further level of validation of the INP parameterizations. Residual characterization enables scientists to measure and evaluate INP concentrations for individual aerosol types even when multiple aerosol sources of INPs are present in the airmass (Cornwell et al., 2019); in the future, such measurements could also be used in case studies to evaluate model predictions of type-dependent INP concentrations, and thereby provide greater insight into reasons for model prediction errors. As a rough guideline, in Table 1 we outline four levels of increasing measurement complexity that can provide increasingly useful information for both closure studies and model evaluation. We also summarize the corresponding level of model evaluation that can potentially be enabled by the measurement suite. In summary, what this articulation of levels of measurement complexity emphasizes is that, while a measurement of INP concentration can enable modelers to answer the question "is the model-simulated INP right?," measurements of aerosol properties are required in order to answer the question "is the simulation right for the right reasons?"

A major advance for the community in recent years has been the pioneering of centralized databases for INP observations by the European BACCHUS project (https://www.bacchus-env.eu/in/). While not comprehensive, this database begins to address the challenge of spatial and temporal representativeness by collecting, in a centralized location, INP measurements from recent and historical field experiments in many parts of the world. It also provides an important first step toward a greater standardization of metadata reporting for INP measurements. Further efforts to collect and, where possible, standardize observational data sets—as has recently been done for measurements of aerosol microphysical and chemical measurements by Reddington et al. (2017)—will accelerate research on the global sources, concentrations, variability, and impacts of INPs.

Finally, more modeling studies are needed that specifically advance INP simulation and evaluate skill in INP simulation as its own problem. By conceptually separating the evaluation of INP simulation from the evaluation of cloud sensitivity responses, we can isolate and solve model biases that may arise from entirely different processes and model errors. To eliminate simulated meteorology as a source of error and isolate a model's aerosol processes for evaluation, modelers can use simulations with either specified dynamics (as in a chemical transport

Table 1
Levels of Measurement Detail for Use in Aerosol-INP Closure Studies and Associated Levels of Model Evaluation Enabled

	Level of measurement detail	Level of model evaluation
Level 0	INP number concentration, reported together with metadata including at a minimum: (1) sampling time, location, and altitude; (2) number of samples; (3) quantitative estimates of the measurement uncertainty; and (4) particle cut-off size or transmission efficiency curve	Evaluate simulated monthly mean INP concentration with INP concentrations from multiple campaigns in different locations. Not possible to distinguish errors in simulation of aerosol amount from errors in parameterization of INP efficiency
Level 1	Level 0 + report a bulk aerosol measurement (mass/total surface area/total number) to provide a basic constraint on simulated aerosol + atmospheric composition information that can aid in validating air mass source (e.g., radon as a tracer for continental air; black carbon as a tracer for combustion sources)	Level 0 + evaluate simulated aerosol for field-campaign-specific time period, using nudged winds or similar methods. Evaluate both model-simulated aerosol and resulting INP concentrations using parameterizations based on total aerosol amount (D10) to disentangle errors in simulation of bulk aerosol from other errors in simulation of INPs. Use accompanying composition data to qualitatively diagnose reasons for observation-model disagreement
Level 2	Level 1 + parallel measurement of the in situ aerosol size distribution that can be used to estimate other moments (number, surface area/mass)	Level 1 + use size distribution information to help diagnose reasons for observation-model disagreement
Level 3	Level 2 + measurement of the size-resolved aerosol composition, ideally on a single-particle basis to allow for assessment of mixing state (e.g., via a single-particle mass spectrometer) or offline composition analysis of filter samples (e.g., by electron microscopy)	Evaluate both the size-resolved composition of model-simulated aerosol and resulting INP concentrations using aerosol-type-based parameterizations to disentangle errors in simulation of aerosol amount, size, and composition from other errors in simulation of INPs. Evaluate aerosol-type-dependent INP parameterizations, when air mass is dominated by a single INP type and other sources are negligible
Level 4	Level 3 + single particle composition measurements of INPs (e.g., via separation using a PCVI)	Level 3 + evaluate the aerosol-type-dependent INP parameterizations in air masses where INPs are contributed by multiple aerosol types

model) or dynamics constrained through methods such as data assimilation or nudging (Sun et al., 2019; K. Zhang et al., 2014) in weather and climate models.

3.4. Understanding and Simulating Cloud and Climate Responses to INPs—Status and Challenges

Regional and global models are the tool of choice for simulating the large-scale weather and climate impacts of INPs, but the multiscale nature of the problem, combined with an incomplete understanding of cloud processes, present challenges for understanding how and when INPs have significant impacts on clouds and climate. In particular, the downstream impacts of INPs on simulated cloud properties are strongly governed by a model's representation of mixed-phase cloud processes. We highlight this challenge in Figure 7, which lists the many processes, crossing multiple scales, that play an important role in cloud responses to INPs and therefore need to be treated in models to fully capture these interactions.

The complexity of cloud ice processes leads to challenges in simulating cloud-climate responses to INPs with confidence. Indeed, in an intercomparison of six GCMs, Storelvmo et al. (2011) found that the response to a particular INP parameterization was quite different in each model, and recent analyses of CMIP5 models have shown that cloud phase responds inconsistently to changes in heterogeneous ice nucleation across different models, with many models producing behaviors that were inconsistent with observational constraints (McCoy et al., 2015, 2016).

The inconsistency of model responses to INPs reveals the importance of a model's resolution, dynamics, and cloud processes, including processes subsequent to ice nucleation, in controlling the simulated cloud response. Several mixed-phase cloud processes that are particularly relevant to cloud responses to INPs, and which are still poorly understood, include secondary ice production (SIP), ice vapor growth, known as the Wegener-Bergeron-Findeisen

(WBF) process, and the treatment of conversions between cloud ice and ice-phase precipitation (Komurcu et al., 2014).

Despite these challenges, regional and global models are currently the primary tools available to advance understanding of large-scale INP-cloud-climate interactions because they remain the only tools capable of simulating the weather and climate impacts of INPs in the broader Earth system. Therefore, here we briefly discuss some of the most important poorly represented and poorly constrained processes that likely limit the ability of current large-scale models to accurately simulate climate responses to changes in INP amount.

3.4.1. Cloud Processes Modulating Responses to INPs and Their Impacts in Mixed-Phase Cloud Systems

Aerosol-ice interactions at the microscale are governed by air parcel cooling rates, which control supersaturation and availability of water vapor; parcel cooling rates in turn are controlled by updraft velocities within the cloud, which are a function of the macroscale cloud dynamics and smaller scale turbulent motions. Thus, the impact of INPs on clouds varies significantly depending on the cloud type (e.g., frontal cloud systems, deep convective anvils, and orographic wave clouds) and the characteristic atmospheric dynamic patterns of the region in which the cloud is located (e.g., typical cloud types differ between mid-latitudes, tropics, and polar regions, and in marine vs. continental air). These physical connections occur across cloud types and have been the subject of many parcel and cloud modeling studies. Many recent studies addressing the mixed-phase cloud regime focus on immersion freezing, on the assumption that in mixed-phase clouds, particles that can serve as INPs are initially activated into cloud droplets. This therefore suggests that mixed-phase clouds are primarily affected by the immersion freezing mode, with deposition freezing playing a minor role, and the role of contact freezing still poorly understood (Ansmann et al., 2008; Kanji et al., 2017). As an example, Simpson et al. (2018) used a parcel model and chamber experiments to investigate how immersion freezing INPs and CCN compete for water vapor in mixed-phase clouds, revealing that the balance of this competition can depend on updraft velocity, INP solubility, and INP size. Since variations in updraft velocity associated with turbulent motions occur at the sub-grid scale in large-scale models, their impacts on cloud microphysics must be parameterized.

Another critical pathway by which INPs impact cloud phase and precipitation is through ice-growth processes, which occur through both vapor deposition and the collection and riming of cloud droplets onto primary ice. In the WBF process, ice crystals grow rapidly via vapor deposition in mixed-phase clouds as a result of the lower equilibrium vapor pressure of ice compared to liquid water, which in turn depletes water vapor, causing cloud droplets to evaporate. While the WBF process is physically well-understood, its representation in large-scale models is a challenge, since they are unable to represent all the relevant scales. For example, GCMs are rarely able to adequately resolve the heterogeneity of hydrometeor phase in simulated clouds and representing cloud phase only as an average across a large grid box can have large impacts on the simulation of the WBF process (Tan & Storelvmo, 2016). A common approach to accounting for sub-grid cloud phase heterogeneity is to apply a constant "correction factor" to the calculated WBF process rate. Simulated cloud phase in GCMs such as CAM5 can be highly sensitive to the value of this correction factor and other model parameters controlling the rate of the WBF process in mixed-phase clouds (Tan & Storelvmo, 2016). More recently, linear eddy simulations with millimeter-scale resolution by Hoffmann (2020) revealed the importance of turbulence and entrainment of dry air for modulating the rate of the WBF process in mixed-phase clouds. Current developments in global cloud modeling, such as multimodeling frameworks and cloud superparameterizations, may offer pathways for the research community to better resolve sub-grid features in the future, but bridging the gap between processes occurring at cloud-resolving scales and at the scale of global climate is still an area of ongoing and active research in modeling of cloud-climate interactions.

Another major source of uncertainty in simulations of the downstream impacts of INPs is the set of processes leading to SIP, which were recently reviewed by Field et al. (2017) and Korolev and Leisner (2020). SIP requires the formation of primary ice via heterogeneous or homogeneous freezing as a prerequisite, though even as few as 10^{-5} to 10^{-3} INPs per L may be enough to trigger SIP (Beard, 1992). SIP processes may be among the most poorly understood areas of cloud microphysics due to the significant challenges in observing these processes in both field and laboratory studies. Current proposed pathways for SIP include rime-splintering, ice-ice collision fragmentation, droplet shattering, and sublimation fragmentation (Field et al., 2017). The rime-splintering process, more commonly known as the Hallett-Mossop process, is often the only SIP pathway represented in

numerical models, commonly using the Cotton et al. (1986) parameterization. This parameterization is dependent on both temperature and riming mass, or the amount of liquid cloud droplets collected onto ice particles, which is prescribed based on the predicted ice particle fall velocities, collection efficiencies, and cloud droplet properties. Modeling studies have investigated how SIP can interact with INP availability to control cloud phase across different types of clouds. For example, increased n_{INPs} resulted in more rimers and greater SIP efficiency in two deep convective cases simulated using a bulk microphysics scheme (Phillips et al., 2007). In contrast, greater n_{INPs} resulted in accelerated glaciation via vapor growth (WBF), which suppressed SIP in regional simulations of wintertime cumulus clouds (Crawford et al., 2012). Modeling studies have also found limited impact of INPs on SIP in convective clouds, where SIP is most strongly impacted by sedimentation or transport of ice particles from aloft (Miltenberger et al., 2020).

Significant advances have been made in modeling and understanding mixed-phase cloud processes through a focus on specific, well-characterized cloud regimes. In particular, single-layer Arctic mixed-phase layer clouds present a unique opportunity to test models of mixed-phase cloud microphysics: they are long-lived and therefore readily observed, they persist under stable conditions with minimal turbulence, and specific cases have been identified where SIP is expected to be minimal, limiting the number of physical processes needing to be observationally constrained (Fridlind et al., 2007). Additionally, large portions of the Arctic are covered by such clouds for a significant fraction of the year, so these clouds are highly impactful for the Earth's radiative budget and climate.

Well-characterized cases of Arctic single-layer mixed-phase clouds have been observed during the1998 First International Satellite Cloud Climatology Project Regional Experiment–Arctic Cloud Experiment/Surface Heat Budget in the Arctic campaign (Curry et al., 2000), the Mixed-Phase Arctic Cloud Experiment (M-PACE; Verlinde et al., 2007), and the indirect and semi-direct aerosol campaign (McFarquhar et al., 2011). Many high-resolution modeling studies (with LES and cloud-resolving models) have used this field campaign as a case study for evaluating and improving understanding of model processes (Avramov & Harrington, 2010; Fan et al., 2009; Fridlind et al., 2007; Klein et al., 2009; Morrison et al., 2005; Paukert & Hoose, 2014; Prenni, Harrington, et al., 2007). As noted previously, one key finding of these studies was the critical role of prognostic simulation of INPs, which produced rapid depletion of INPs from the cloud layer, enabling simulated clouds to be sustained longer (Avramov & Harrington, 2010; Fan et al., 2009; Fridlind et al., 2007; Harrington & Olsson, 2001; Morrison et al., 2005, 2011; Prenni, Harrington, et al., 2007). An intercomparison of single-column models and cloud-resolving models on the basis of the M-PACE experiment suggested that the models' representations of mixed-phase microphysics played a key role in their ability to match the observations, with models having more sophisticated microphysics schemes performing somewhat better in their simulation of cloud liquid and ice water path (Klein et al., 2009; Morrison et al., 2009).

Increasingly, careful observational analyses have also been able to shed light on the emergent responses of cloud systems to perturbations in INP concentrations. For example, a recent analysis of cloud and aerosol properties from satellite retrievals showed that cloud optical thickness, cloud thickness, and cloud fraction of large-scale ice cloud systems (e.g., clouds associated with frontal systems) increase with aerosol loading monotonically, whereas these properties of convective-generated ice clouds (i.e., anvil ice clouds) first increase and then decrease with increasing aerosol loading (Zhao et al., 2018).

3.4.2. Cirrus Cloud Responses to INPs and Proposals for Geoengineering Through Cirrus Cloud Thinning

Cloud dynamics also control aerosol-ice interactions in cirrus clouds, where the competition between homogeneous and heterogeneous freezing is largely controlled by updraft velocities (DeMott et al., 1997; Kärcher & Voigt, 2006; Sassen & Benson, 2000). In the tropical tropopause layer, this competition is controlled by small-scale variations in updraft velocities caused by gravity waves (Jensen et al., 2010; Shi & Liu, 2016; Y. Wang et al., 2014). For heterogeneous freezing in the cirrus cloud regime, mineral dust particles are the dominant source of INPs, and freezing may occur either via the immersion freezing or the deposition freezing pathway (Cziczo et al., 2013).

Since cloud properties depend not only on INPs, but are also highly influenced by other factors including the dynamic and thermodynamic environment, and the history of the cloud's development, it is desirable to identify

targets where the number of processes contributing to controlling the cloud properties can be minimized and individual processes more effectively isolated. Orographic wave clouds have been leveraged in this regard, since they present a unique case where flow is relatively laminar, air parcels follow predictable streamlines, and their residence time in the cloud is limited to a few minutes. For this reason, observations of cloud ice can be more directly connected to upstream observations of INPs. This was done, for example, in the Ice in Clouds Experiment–Layer Clouds study (Eidhammer et al., 2010; Field et al., 2012), where it was shown that aerosol size distribution and composition measurements could be used to successfully predict primary ice formation in this well-constrained case.

Understanding the impact of INPs on cirrus clouds has also been a focus of recent geoengineering proposals. Unlike most other cloud types, cirrus clouds exert a net warming effect on climate. "Cirrus cloud thinning" (CCT; Lawrence et al., 2018; Mitchell & Finnegan, 2009; Storelvmo et al., 2013) is a climate intervention approach that aims to reduce the thickness and extent of cirrus clouds, counteracting their warming effect.

In CCT, INPs are injected or "seeded" into cirrus clouds to enable heterogeneous ice nucleation before the onset of homogeneous ice nucleation. When INPs are unavailable to cirrus clouds, homogeneous freezing can occur, freezing most or all of the cloud's supercooled droplets. In contrast, if INPs are injected as part of CCT interventions, only a subset of droplets would freeze—via heterogeneous freezing on the injected particles. These crystals would grow rapidly, producing larger ice crystals that sediment more rapidly compared to the unperturbed cloud. In this way, CCT aims to deplete and thin cirrus clouds and so may be effective in extratropical and polar regions where homogeneous freezing makes up a significant fraction of naturally occurring ice formation in cirrus clouds (Storelvmo & Herger, 2014; Storelvmo et al., 2013).

However, modeling studies disagree on the effectiveness of CCT strategies, with some studies suggesting it may be ineffective (Gasparini & Lohmann, 2016; Penner et al., 2015) and others warning of a risk of overseeding, where INP injections in cloud-free regions could increase cirrus cloud formation and unintentionally increase warming (Storelvmo & Herger, 2014; Storelvmo et al., 2013).

4. Summary and Conclusions

With the recent growth of research focused on the measurement and characterization of INPs (Coluzza et al., 2017; DeMott et al., 2011; DeMott, Möhler, et al., 2018; Hoose & Möhler, 2012; Kanji et al., 2017; Murray et al., 2012), we believe the research community stands on the cusp of significant breakthroughs in advancing predictive understanding of INPs, and in representing the processes that control their temporal and geographic variability in atmospheric models. However, further work is still needed to fully incorporate recent experimental findings into atmospheric models, and evaluate model predictive skill for n_{INPs}. Continued work is also needed to develop observational data products that advance fundamental understanding, enable a richer representation of relevant processes in models, and allow for a more systematic evaluation of model realism.

We have summarized the recent literature on cloud responses to INPs, in particular their impacts on climate. Recent intercomparisons show that GCMs continue to disagree on the strength, nature, and in some cases the sign of cloud responses and climate feedbacks to INPs; this is true for responses in both mixed-phase and cirrus clouds. Many of the cloud microphysical processes controlling cloud responses to INPs continue to be poorly understood or challenging to parameterize adequately in large-scale models. Given these complexities, most recent regional and global modeling studies exploring the impact of INP on clouds have actually answered the question "how sensitive are simulated clouds or climate in a specific model to changes in INPs?" and not the question "how sensitive are clouds or climate to INPs in the real world?"

The response of clouds to INPs may be better simulated by high-resolution models, but these models are unable to capture interactions with the broader climate and Earth system. Bridging the gap in scales between cloud microphysical and macrophysical properties, and the simulation of regional- and global-scale cloud feedbacks continues to present a challenge to the research community. While this gap remains, as was previously emphasized by Kanji et al. (2017), modeling studies should be thoughtfully designed at a scale appropriate to the science question of interest.

With these considerations in mind, here we briefly summarize key research needs, identified throughout this paper, which will help to bridge the observation-model gap for INP prediction and advance the field in this area.

1. **Careful design of laboratory measurements of ice nucleation is key to the development of parameterizations appropriate for use in modeling studies** (Section 2). A key barrier in representing ice nucleation in models continues to be the lack of a readily available comprehensive suite of parameterizations, potentially using a common theoretical underpinning, that covers the entire relevant temperature and humidity range as well as all relevant particle types. We encourage experiments to fulfill the four criteria for parameterization development and emphasize the importance of clearly defining experimental and parameterization limitations to help guide decisions in model implementation.
2. **Explicit treatment of time-dependent behavior is of second-order importance for INP predictability in regional and global atmospheric models** (Section 2.1 and Section 3.1.1). Therefore, when addressing the problem of bridging the model-observation gap, the relevance of time dependence is primarily due to its potential impact on values of J_{het} and n_s that are inferred from experimental results. Experimentalists should consider under which conditions it may be appropriate to either account for time dependence explicitly, or correct for its effects, when deriving parameterizations of INP efficiency from experiments. Additionally, small-scale kinetic effects, such as the responses of freezing processes to variations in updraft velocities associated with in-cloud turbulence, may be sensitive to the treatment of time dependence. These effects should be studied using laboratory studies and small-scale models (e.g., direct numerical simulations and parcel models), so that their net effects can be parameterized inside of large-scale models, where significant.
3. **Characterization of particle size distributions and the contributions of supermicron particles should be emphasized in field and laboratory experiments** (Section 2.2). Particle size is one of the critical parameters governing the ice nucleation behavior of particle populations, but is not always well-characterized in experiments. We emphasize the need for experiments to include accurate measurements of the particle size distribution and the surface area participating in ice nucleation. Additionally, supermicron particles are systematically undersampled, and more experiments are needed that specifically quantify their contributions to atmospheric INPs.
4. **Size-resolved measurements of aerosol and INP chemistry are required to test the limits of INP predictability on the basis of observed aerosol in carefully designed closure experiments** (Section 2.3). Together with size, particle chemistry is the second critical parameter that govern aerosol particles' efficiency as INPs. In complex aerosol populations, both quantities must be measured simultaneously, that is, aerosol chemistry as a function of particle size, to enable a strong test of the applicability of parameterizations through closure experiments. When possible, we encourage such experiments to be conducted in tandem with direct characterization of ice crystal residuals; this combination provides the strongest possible test of INP predictability on the basis of observed aerosol properties.
5. **A larger emphasis should be placed on the development of ice nucleation parameterizations that are representative in the real environment, and on testing parameterizations against atmospherically relevant observations not used in their development** (Section 3.1). Ongoing efforts in this direction address the potential for experimental factors to limit the representativeness of existing parameterizations for atmospheric INPs, for example, due to sample processing methods (e.g., milling or sieving), instrument limitations (e.g., unavoidable losses of supermicron particles), and aerosolization processes that differ from those in the real environment.
6. **Continued work is needed to further advance the development and observational testing of emission parameterizations for key aerosol types contributing to INPs that are rarely represented in atmospheric models** (Section 3.2). Model representations of particle source functions for certain key aerosol types, including several classes of biological particles and agricultural soil dusts, are either lacking or still in the early stages of development. However, such parameterizations are required if INP sources, their dependence on changes in both climate and land use, and resulting climate impacts and feedbacks, are to be simulated in Earth System models. The further development of such parameterizations, and their evaluation against appropriate observational data sets that are independent of those used in developing the parameterizations, remains a priority.
7. **Measurement and modeling of the vertical profiles of aerosol and INPs, and their concentrations remote from source regions, represents a critical need** (Section 3.2.2). With a growing use of aerosol-aware ice nucleation parameterizations, evaluation of simulated aerosol must go beyond evaluation of

column-integrated quantities (e.g., aerosol optical depth) to assess aerosol vertical distributions and aerosol number concentrations remote from source regions. Additionally, most regional and global models do not currently have a "prognostic INP" capability that distinguishes the impacts of cloud processes (e.g., activation and removal) on the INP activity of the existing aerosol population, which may lead to additional biases. Simulation of aerosol in Earth System Models suffers from greater biases at high altitudes and in remote regions, due to the cumulative impacts of errors in the simulation of parameterized vertical transport (due to turbulence and convection) and removal processes (wet scavenging and dry deposition). Simultaneously, higher altitudes, and remote high latitude regions, are the locations where INP-cloud interactions and the resulting feedbacks on climate system are expected to be largest. Observational and modeling studies that target and constrain processes controlling transport to these regions remain a research priority.

8. **Increased observational detail in field experiments through measurement of detailed aerosol properties, increased spatial and temporal coverage, and closure experiments** (Section 3.3, Table 1). While many studies have taken a bottom-up approach toward understanding INPs, by developing parameterizations from laboratory experiments on individual particle types, relatively fewer studies to date have taken the top-down view, by using ambient measurements to assess whether closure can be achieved between the predicted and measured ambient INP amount across a variety of land use types and environments. As such, we highlight that the potential impact of observational data on our scientific understanding can be maximized with increased measurement detail (Table 1), increased measurement duration and spatial coverage, and carefully designed observational closure studies.
9. **The aggregation of observational data into community databases and the development of common metadata and measurement standards have the potential to accelerate scientific insights gained from the analysis of globally collected INP measurements** (Section 3.3). The recent development of an observational database of INP measurements by the BACCHUS project represents a significant service to the field. Such efforts make observations more accessible to a broader cross-section of the scientific community for a variety of efforts, including evaluation of the skillful simulation of INPs in models. Greater standardization of reported metadata and measurement approaches for the observations contributed to such databases will further reduce barriers to their use in model development and evaluation.
10. **Additional studies are needed that evaluate the skill of regional and global models in simulating INPs together with their particle sources** (Section 3.3). As models incorporate aerosol-aware representations of cloud ice processes, those processes become sensitive to the models' skill and biases in prediction of INPs. However, due to the historically limited availability of observational constraints from field studies, regional and global models often have incorporated INP effects on clouds after only very limited evaluation of the accuracy of simulated INP concentrations in the model. As more measurements from field observations become available, further studies are needed that specifically characterize these biases and offer insight into their causes through parallel evaluation of simulated aerosol and related atmospheric processes.
11. **Cloud regime-specific modeling studies are needed that quantify cloud sensitivities to INPs, which in turn will provide regionally specific guidance on the accuracy with which INPs need to be measured and modeled** (Section 3.4). Efforts to observe and simulate INP quantities ideally should prioritize those particle sources that have an important impact on cloud processes and climate at regional to global scales. However, the complexities in modeling cloud responses to INPs have limited the clarity with which modeling studies can provide guidance on the sensitivity of cloud responses. Historically, significant progress has been made in understanding and modeling cloud ice processes through focused efforts on the simulation of cloud responses in well-characterized case studies of specific cloud regimes that were selected due to their simpler microphysics (e.g., Arctic cirrus, orographic wave clouds). These studies included intercomparisons of models that represented processes, and at different scales (from LES to global). More studies are needed with models and case studies of additional processes that play a key role in controlling cloud-climate feedbacks and their sensitivity to INPs. In particular, the WBF process and other SIP processes play a key role in controlling the responses of mixed-phase clouds to INPs, and the fidelity of their parameterization across models of different scales represents a key research target.

Addressing these key research needs will require the efforts of a broad community including experts in ice nucleation measurements, aerosol measurements and characterization, particle surface chemistry measurements, aerosol process modeling, high-resolution cloud modeling, and global modeling of aerosol-cloud-climate feedbacks. In closing, therefore, we would like to advocate not only for separate efforts, but also for collaborative

efforts between the modeling and measurement communities to tackle these challenges through a joint measurements-to-models approach.

Data Availability Statement

Data were not used, nor created for this research. Data underlying the figures reproduced herein are available through the original articles from which they are reproduced: Worringen et al. (2015), China et al. (2017), B. Wang et al. (2016), and Cornwell, Xiao, et al. (2021).

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