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REVIEW OF RECENT DEVELOPMENTS AND SHORTCOMINGS IN THE CHARACTERIZATION OF POTENTIAL ATMOSPHERIC ICE NUCLEI: FOCUS ON THE TROPICS

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Abstract

In this paper, we summarize the four main ice nucleating aerosol types: mineral dusts, bioaerosols, soot, and glassy organics, with the aim of demonstrating the limitations in scientific literature regarding their ice nucleation properties. Because the tropics are largely associated with marine environments, such as the Atlantic, Pacific, and Indian Oceans, they are potential source of ice nuclei, and therefore ice clouds can form in regions of high convective uplift. As a result, these particles are able to influence both the planet's radiative force and its hydrological cycle. Due to our limited understanding of these effects, we would like to encourage the scientific community to increase its efforts to study and characterize the tropical aerosol particles that may function as ice nuclei. Through such efforts, we may reduce uncertainties in climate predictions, and improve our understanding of global warming in the hopes of finding potential solutions to these issues.

Keywords: Ice nuclei, Ice clouds, aerosols, climate change.

Introduction

Ice clouds have a significant impact on the Earth's energy budget and hydrological cycle (Ramanathan *et al.*, 2001; Baker and Peter, 2008; DeMott *et al.*, 2010). However, their impact on climate represents one of the largest uncertainties in the forecasting of future climate (Forster *et al.*, 2007). As shown in Figure 1, clouds affect the radiative properties of the Earth by trapping its outgoing infrared radiation and reflecting incoming visible solar radiation, thus having both a warming and cooling effect on the Earth, respectively (Baker and Peter, 2008). Additionally, because between 60% and 70% of the total precipitation is initiated by ice water clouds, these clouds substantially control the water exchange between the ocean and continents as well as between the planetary surface and the atmosphere (Lohmann and Diehl, 2006). Although most of the precipitation in the tropics is produced by warm clouds, ice clouds are responsible for 20% to 30% of the precipitation at tropical latitudes (Lau and Wu, 2003). Tropospheric ice clouds can be divided into two main categories: cirrus and mixed phase clouds. While cirrus clouds are composed of ice crystals and exist in the upper troposphere, mixed phase clouds are composed of both ice crystals and water droplets, and exist in the low and middle troposphere (Hartmann *et al*., 1992). Due to their low altitude, mixed phase clouds have an important influence on the hydrological cycle by increasing precipitation over the ocean and continents (Quante, 2004).

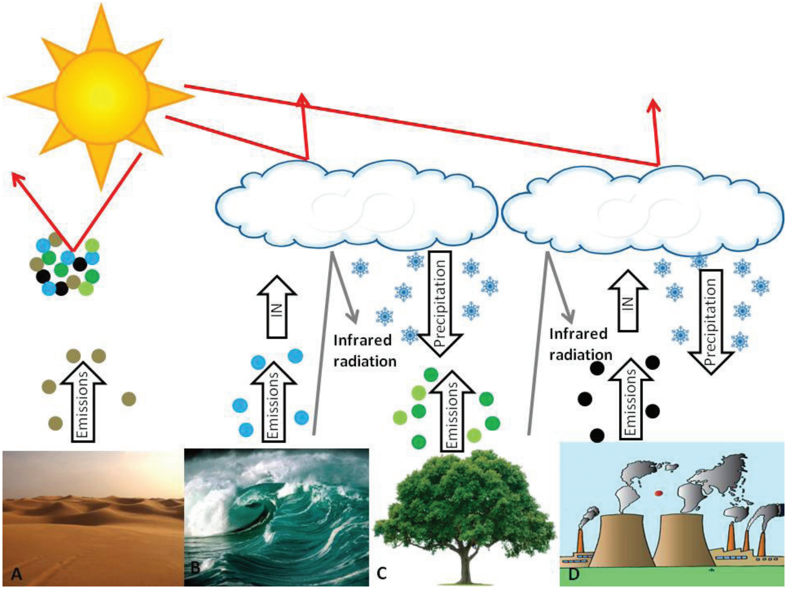

Figure 1. *Schematic of the direct and indirect effect of anthropogenic aerosol particles. The potential ice nuclei (IN) sources are also illustrated: A) Deserts (mineral dust), B) Oceans (marine aerosols and sea salt), C) Forests (bioaerosols and organic aerosols), D) Industry and biomass burning (soot).*

Ice crystals form through both homogeneous and heterogeneous ice nucleation at temperatures less than and greater than -38 °C, respectively. While homogeneous freezing is important for cirrus cloud formation, heterogeneous freezing is the dominant process by which mixed-phase clouds form. Heterogeneous ice nucleation requires the presence of ice nuclei (IN), which generally represents one in 10^5 ambient particles (DeMott *et al*., 2010). Heterogeneous ice nucleation occurs by four different modes:

1. Deposition nucleation, where water vapor deposits directly onto an IN as ice.
2. Condensation freezing, where liquid water condenses on IN to form a liquid droplet at temperatures where quickly freezes.
3. Immersion freezing, where an IN is immersed in a liquid droplet, and eventually freezes upon cooling.
4. Contact freezing, in which IN collide with super-cooled liquid droplets to cause ice nucleation.

In addition to the dependence of the nucleation efficiency of these four modes on temperature and relative humidity, there is also a dependence on the physical and chemical properties of the surfaces of the ice nuclei that facilitates ice embryo formation (Pruppacher and Klett, 1997; DeMott *et al*., 2011).

As mentioned above, ice nuclei in clouds that are warmer than -38 ^{o}C represent fewer than about 1 in 10^5 aerosol particles (DeMott *et al*., 2010). This low number concentration demonstrates the selective characteristics of ice nuclei. Pruppacher and Klett (1997) discussed what are considered to be the most important of these, five of which are summarized below:

1. Physical state: Ice nuclei are generally water-insoluble as a solid substrate is required for ice embryo formation. Soluble substances such as salt cause freezing point depression.
2. Particle size: Aerosols larger than the Aitken size range (0.01 - 0.01 μm) have been reported to be more efficient ice nuclei than Aitken particles. The size of efficient ice nuclei has been found to depend on the chemical composition of the aerosol as well as the environment's supersaturation. In addition, because smaller particles have a greater solubility, it is not surprising that ice nucleation tends to occur on larger aerosols. Therefore, particles in the Aitken size range may function as ice nuclei if they are highly water-insoluble.
3. Hydrogen bonding: Because ice is held together by hydrogen bonds, ice nuclei must possess the ability to form hydrogen bonds with water. In addition, the hydrogen-bonding molecules on the surface of the ice nuclei must possess rotational symmetry so that the hydrogen-bonding groups are always exposed (Fukuta, 1966).
4. Crystallographic arrangement: The geometric arrangement of hydrogen bonding molecules on the surface of the ice nucleus must be very close to that of water molecules in an ice crystal lattice. Crystallographic differences will result in strain at the ice-substrate interface, which will reduce the efficiency of the ice nucleus.
5. Active sites: It has been found that ice nucleation occurs on distinct active sites on substrates. Active sites typically refer to surface defects (growth steps, cracks, cavities) and chemical impurities at a solid's surface, among other physical characteristics. Consequently, not all aerosol particles having the favourable chemical composition and crystallographic structure, as were previously described, are equally efficient ice nuclei.

As can be seen above, both the modes of ice nucleation and the theoretical requirements of efficient ice nuclei have been identified and described extensively. Although the scientific community has made a great effort to understand the ice cloud formation mechanisms in the laboratory, there is a lack of information from field studies (e.g. Hoose and Möhler, 2012). This deficiency is more pronounced in the tropics even though cloud formation, and hence precipitation, plays an important role in agriculture, which is a very important component of the local economies. Precipitation could also potentially impact the tourism, transportation and natural ecosystems among other local activities. The tropics are also characterized by a large amount of convective uplift that takes air, much of it marine, to the upper troposphere where cirrus clouds may form. The Hadley Cell then transports this air to high latitudes, prior to descending.

Additionally, there is still a lack of understanding when it comes to describing the results of ice nucleation experiments aiming to characterize potential ice nuclei. Because previous reports are insufficient to draw quantitative conclusions regarding the ice nucleation abilities of various atmospheric aerosols, further investigation is required in each of the four nucleation modes, as will be demonstrated in this paper. This manuscript also introduces the great potential that the tropics have as an important source of efficient IN due to the presence of different biomes.

Ice nucleation efficiency of particles of current interest

A wide variety of particles have been examined and characterized in terms of their ice nucleation abilities in each of the four nucleation modes, which are deposition nucleation, condensation freezing, immersion freezing, and contact freezing. As illustrated in Figure 1, four of the more common ice nuclei categories are:

i. Mineral dusts
ii. Bio-aerosols
iii. Carbonaceous combustion material or soot
iv. Glassy organic aerosols

Each of these categories will be described and examples will be given of the temperatures or saturations with respect to ice that required for them to initiate ice nucleation in each freezing mode.

2 Mineral dusts

Approximately 2000 Tg of mineral dust is aerosolized per year (Jaenicke *et al.*, 2007), half of which may be related to anthropogenic activity that includes desertification and deforestation. One of the most notable sources of mineral dust is the Dust Belt which extends from the west coast of North Africa into Central Asia (Prospero, 2002), part of which is located in tropical latitudes. The maximum average surface concentration of mineral dust particles has been found to be 65 cm^{-3}, which only represents a small fraction of total aerosol particles, which are present in the atmosphere at a concentration of 100,000 cm^{-3} (Hoose *et al.*, 2010; Jaenicke *et al.*, 2007). It has been found that between roughly one third (in mixed-phase clouds) and one half (in cirrus clouds) of ice nuclei are composed of mineral dust (DeMott *et al.*, 2003; Twohy & Poellot, 2005; Richardson *et al.*, 2007; Pratt *et al.*, 2009). In addition, ice nucleation simulations conducted by Hoose *et al.* (2010) concluded that mineral dusts account for approximately 77% of ice nuclei. Mineral dusts are detected and identified in the atmosphere using their various crystallographic structures, particle morphologies, and compositions by an assortment of techniques including X-ray Diffraction, electron microscopy, and mass spectrometry, respectively (Murray *et al.*, 2012).

Minerals are crystalline solids having a specific chemical composition and structure that occur naturally in the environment. The most frequently observed mineral group in the atmosphere are clays, most of which are composed of a layered structure of silicon dioxide tetrahedrals and aluminum oxide octahedrals (Yakobi-Hancock *et al.*, 2013). Clays that have been examined in ice nucleation studies include kaolinite (Lüönd *et al.*, 2010; Ladino *et al.*, 2011), illite (Hoffer, 1961), and montmorillonite (Conen *et al.*, 2011). Arizona Test Dust (ATD), NX illite and Mojave Desert Dust (MDD) are some examples of laboratory proxies that have been used for natural mineral dust as they have been well characterized with respect to particle size, surface area, and mineralogy in the past (Broadley *et al.*, 2012; Ladino and Abbatt, 2013; Yakobi-Hancock *et al.*, 2013).

Mineral dusts have been identified as relatively efficient ice nuclei by many older studies, largely due to their crystallographic match with ice and ability to form hydrogen bonds at the surface. For example, Hoffer (1961) identified the median freezing temperatures of heterogeneous immersion freezing on kaolinite, montmorillonite, and illite to be -32.5 °C, -24.0 °C, and -23.5 °C, respectively. As was discussed previously, one important characteristic of ice nuclei is the presence of active sites. Therefore, active site densities can be correlated to the ability of an aerosol to function as an ice nucleus. In other words, ice nuclei with more active sites exhibit greater ice nucleation efficiencies. For this reason Murray *et al.* (2012) compiled and plotted several recent data sets obtained from immersion ice nucleation studies that were conducted using mineral dusts that originated from, or were relevant to, arid regions. This comparison is shown in Figure 2 below.

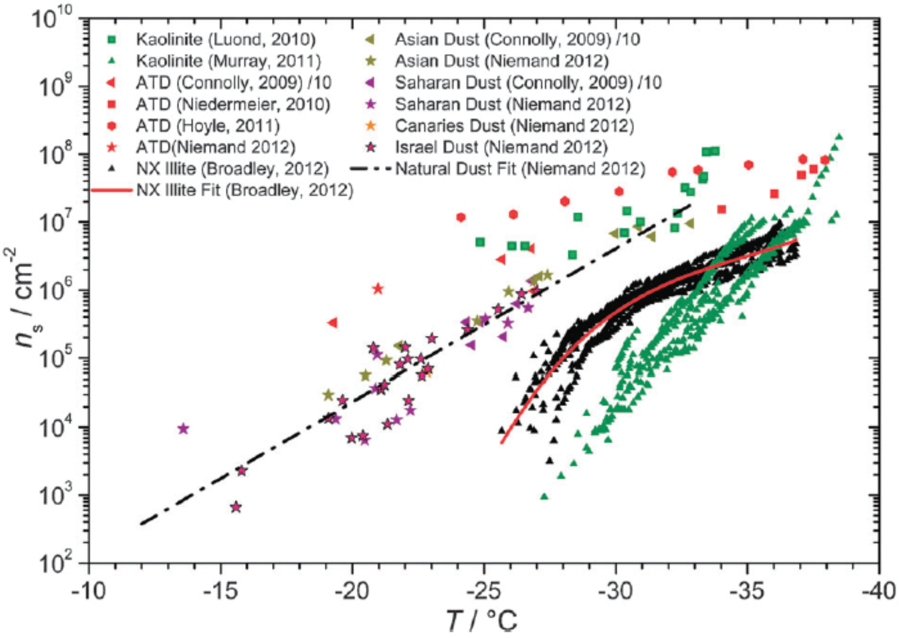

Figure 2. *Compiled mineral dust active site densities (n_s, cm^{-2}) for immersion freezing between -10 °C and -40 °C from Murray et al. (2012).*

According to Figure 2, mineral dusts would exhibit active site densities of greater than 10^2/cm^2 at temperatures as warm as -10 °C, which is about 28 °C warmer than the freezing temperature of pure water. Because of this it can be said that mineral dusts are efficient ice nuclei. It should also be noted from this plot that Arizona Test dust (ATD) and Saharan Dust have been identified as the more efficient mineral dust ice nuclei, while kaolinite and the proxy NX illite are relatively inefficient. As a result of this inefficiency atmospheric ice nucleation cannot be fully explained by mineral dusts, which one of the motivations for other potential sources to be explored.

Deposition and condensation ice nucleation has also been studied extensively. Archuleta *et al.* (2005) found that 200 nm mineral dust particles initiated ice formation in the deposition mode at an ice relative humidity of about 135%, regardless of temperature. Bundke *et al.* (2008) found that there was a sharp increase in the activation ratio at temperatures less than -16 °C and ice supersaturation of greater than 20%. Connolly *et al.* (2009) found that Arizona Test Dust exhibited strong deposition mode nucleation at temperatures less than -24 °C, while the freezing efficiency of Sahara Dust increased between -24 °C and -27 °C. Yakobi-Hancock *et al.* (2013) found the critical supersaturation of feldspar required to nucleate ice through deposition nucleation is 128% at -40 °C, which is much lower than the other tested metal oxides and minerals. Connolly *et al.* (2009) hypothesized that other processes, such as contact nucleation, may result in ice particle formation at warmer temperatures. For instance, a comparison of the frozen fractions from contact and immersion freezing on kaolinite particles as a function of temperature was made by several authors, as reviewed by Ladino *et al.*, (2013). It was found that the same aerosol particles of similar sizes were able to nucleate ice at warmer (5-9 °C) temperatures through contact freezing as compared to immersion freezing.

3 Bioaerosols

Primary biological aerosol particles (PBAPs) refer to all atmospheric particles that originated from living organisms (Gabey *et al.*, 2010). These particles include fragmented plants and animals, pollen, fungal spores, bacteria, and virus (Deguillaume *et al.*, 2008). It has been found that approximately 1000 Tgyear^{-1} of these particles enter the atmosphere, making this source comparable to major sources such as mineral dust (2000 Tgyear^{-1} and sea salt (3290 Tgyear^{-1}) (Jaenicke *et al.* 2007). Using Aerosol Time of Flight Mass Spectrometry (ATOFMS), Pratt *et al.* (2009) demonstrated that cloud-ice crystal residues are comprised of about 33% biological material and 50% mineral dust, indicating that biological particles are an important component of ice nucleation processes. Möhler *et al.* (2007) discussed some of the most common PBAPs, which includes bacteria, pollen, fungal spores, and marine material, and are reviewed below.

3.1 Bacteria

The average atmospheric number densities of bacteria over continents have been found to be about 10^{-2} to 10^{-1} cm^{-3} (Hoose *et al.*, 2010). Although this concentration is relatively low in comparison to that of mineral dust particles (65 cm^{-3}) the ice nucleating properties of bacteria have been extensively studied due to their initiation of frost injury of plants through ice nucleation (Mohler *et al.* 2007; Morris *et al.* 2008, 2013; Kanji *et al.* 2011; Garcia *et al*. 2012; Attard *et al.* 2012; Goncalves *et al*. 2012). This ability is due to genes that encode for an ice nucleation protein that is located in the bacterial cell membrane (Graether and Jia, 2001). This protein initiates ice nucleation by orienting the water molecules into an ice-like crystallographic structure (Morris *et al.*, 2004).

The bacterial strain *Pseudomonas syringae* can be found on the surfaces of plant leaves and is one of the most abundant, well-known ice nucleation-active (INA) bacteria currently known. It is also one of the most efficient ice nuclei as it is able to cause immersion heterogeneous freezing at temperatures as warm as -2 ^oC (Gross *et al.*, 1983; Lindemann *et al.*, 1982). However, relatively low ice nucleation frequencies were observed as only 1 out of 10,000 cells express the ice nucleation phenotype (Maki *et al*., 1974).

In addition to immersion freezing, deposition nucleation has also been observed and tested using Snomax^{TM}, which is a proxy for biological ice nuclei composed of proteins originating from *Pseudomonas syringae* (Hoose and Möhler, 2012). Snomax^{TM} is commoly use for the artificial production of snow due to its high IN efficiencies. Chernoff & Bertram (2010) found that it is a fairly efficient ice nucleus as it exhibited onset ice supersaturations between 110% and 120%.

3.2 Pollen

It has been estimated that the average continental atmospheric number densities of pollen are approximately 10^{-6} to 10^{-5} cm^{-3}, and are subject to a large seasonal variability (Hoose *et al.*, 2010). Pummer *et al.* (2012) examined the immersion ice nucleation characteristics of various birch and conifer pollens. The reported median freezing temperatures were between -19 ^oC and -33 ^oC. These temperatures were attributed to the presence of polysaccharides that are commonly found on the surfaces of pollen and have been found to cause ice nucleation in past studies (Goldstein and Nobel, 1991). When investigating deposition mode ice nucleation at temperatures down to -33 ^oC and super-saturations with respect to ice up to 35%, Diehl *et al.* (2001) found that while the pollen was able to absorb water from the humid environment due to the capillary effect, they did not function as deposition ice nuclei. However, this study also found that their mean condensation freezing efficiency of 50% was reached at temperatures that ranged from -12 ^oC to -18 ^oC. In a subsequent investigation of immersion freezing on pollen, Diehl *et al.* (2002) found that activation occurred at temperatures up to -9 ^oC, while contact freezing occurred at temperatures up to -5 ^oC.

The discrepancy between the immersion freezing results of Pummer *et al.* (2012) and Diehl et al (2002) is an example of the lack of understanding regarding pollen as ice nuclei. Because of this, more quantitative studies are required in order to better understand the mechanism by which pollen functions as an ice nucleus in the four different modes.

3.3 Fungal spores

Spores, or the reproductive units of fungi, are released into the air and scattered throughout the atmosphere by air currents (Iannone *et al.*, 2011). Elbert *et al.* (2007) found that the average continental number concentration of fungal spores is between 10^{-3} and 10^{-2} cm^{-3}, which is comparable to atmospheric bacterial concentrations. Iannone *et al.* (2011) studied the immersion ice nucleation properties of one of the most abundant types of spores found in the atmosphere, *Cladosporium*, which composes approximately 40% of the total spore count (Li and Kendrick, 1995; Al-subai, 2002; Herrero *et al.*, 2006). It was found that the spores were activated at -28 ^{o}C, indicating that in comparison with other biological material this genus is an inefficient ice nucleus. This was attributed to a hydrophobic protein coating that is common on the surface of filamentous fungi such as *Cladosporium* (Iannone *et al.* 2011). Jayaweera and Flanagan (1982) found that fungal spores found in the Arctic atmosphere such as *Penicillium digitatum* initiated immersion freezing at temperatures as high as -10 ^{o}C.

3.4 Marine material

Much of the research regarding biological ice nuclei has been focused on continental aerosols such as the previously mentioned bacteria and pollen. However, observed atmospheric ice nuclei levels above remote marine environments has been found to be caused by marine biological activity and the subsequent production of bubble bursting and therefore sea spray production (Blanchard 1989). For example, Bigg (1973) found that elevated atmospheric immersion ice nuclei concentrations have been associated with phytoplankton blooms. It was also demonstrated that the mean number concentrations of ice nuclei at -15 ^{o}C were as high as 58 m^{-3} above the Antarctic Ocean (Bigg 1973). Activation temperatures in the immersion mode have been found to be as high as -3 ^{o}C for plankton and -7 ^{o}C for various algae species (Lundheim, 1997; Schnell, 1975).

Knopf *et al.* (2010) examined diatoms, which are microalgae having siliceous cell walls in a matrix of proteins and polysaccharides, that are extremely abundant in oceanic water having a high degree of nutrient upwelling and therefore productivity (Alvain *et al.*, 2008). It is thought that the surface oriented hydrophilic functional groups of the organic matrix provide a structural match with ice (Knopf *et al.*, 2010). It was found that diatoms initiated deposition mode and immersion mode ice nucleation at temperatures less than and greater than -33 ^{o}C, respectively (Knopf *et al.* 2010). Alpert *et al.* (2011) reported that the diatom *N. atomus* is capable of initiating deposition ice nucleation at temperatures below -33 ^{o}C and relative humidity with respect to ice that are greater than 121%.

To summarize, the ability of biological particles to act as ice nuclei, and therefore facilitate cloud formation and increase precipitation, depends on the atmospheric concentrations of active ice nuclei species. Because many of these concentrations vary temporally and spatially, these effects vary with the seasons and location. Therefore, in order to better understand their effects on clouds and climate more quantitative observations of active biological ice nuclei are required (Després *et al.*, 2012). Large concentrations of biological material can be found in the tropics due to the extensive wooded areas; however this has been poorly quantified.

4 Carbonaceous combustion aerosols (Soot)

Soot particles are solid, carbonaceous products resulting from the incomplete combustion of carbon containing material such as fossil fuels and natural biomass (Chou *et al.*, 2013). Approximately 8.2 Tg of black carbon or soot is emitted globally per year (Ito and Penner, 2005). Through the use of an aerosol mass spectrometer (AMS), scanning mobility particle size (SMPS), and ATOFMS, Cozic *et al.* (2008) identified soot particles in ice crystals, indicating that they are capable of functioning as ice nuclei. In addition, it was found that while the black carbon mass fraction comprised only 5% of the total aerosol, it was enriched to about 27% of the ice residue, which further suggests that black carbon possesses ice nucleation characteristics (Cozic et al 2008). In contrast, by characterizing the elemental compositions of residual ice nuclei from the Amazon basin using transmission electron microscopy with energy-dispersive X-ray spectroscopy (TEM-EDX), Prenni *et al.* (2009) found that soot particles were not present, which suggested that biomass-burning particles do not function as ice nuclei in the atmosphere.

While some studies have found that soot particles function as relatively good ice nuclei, others have demonstrated that this is not the case. Popovicheva *et al.* (2008) found that soot having a high degree of porosity behaved as efficient ice nuclei in the immersion freezing mode. Similarly, Gorbunov *et al.* (2001) argued that soot particles are a very potent source of ice nuclei. That can explain the ice crystal number concentrations observed in low clouds. Corbin *et al.* (2012) found that biomass burning particles behaved as ice nuclei as they initiated deposition nucleation at 134% RH_i and -34.2 ^{o}C.

In contrast, Koehler *et al.* (2009) found that hygroscopic soot particles did not nucleate ice through deposition or condensation modes at temperatures warmer than that of homogeneous freezing. DeMott *et al.* (1999) found that soot particles functioned as ice nuclei at an onset temperature of -24 ^{o}C during immersion freezing. Similarly, Diehl and Mitra (1998) found that freezing was initiated between temperatures of -18 ^{o}C and -28 ^{o}C in the immersion mode. Fornea *et al.* (2009) found that soot was not activated in the contact freezing mode until a temperature of -25.6 ^{o}C was reached.

The variation in the ice nucleation properties of soot particles may be attributed to the variation in their aging processes, the fuel that produces them, and their mixing states. These in turn have been found to affect the particles' chemical compositions, sizes, shapes, and therefore their ice nucleation properties (Andreae and Gelencsér, 2006; Riemer *et al.*, 2009; Buseck *et al.*, 2012). For example, Weingartner *et al.* (1997) found that freshly emitted soot particles are mainly hydrophobic, which reduces their ability to function as efficient ice nuclei. However, atmospheric aging processes have been found to coat black carbon with secondary aerosol material such as inorganic ions (Weingartner *et al.*, 1997). These coatings allow black carbon to exhibit a more hygroscopic character, and may therefore allow it to function as a better ice nucleus. In addition, aggregates of soot have been found to fuse together to form porous agglomerates (Popovicheva *et al.*, 2008). Such pores may function as active sites to increase the ice nucleation efficiency of black carbon. In contrast, while soot is porous, it is also amorphous, and therefore may lack the ice-matching crystalline structure required for ice nucleation (Buseck *et al.* 2012).

Although most lab experiments have shown that soot behaves as a good ice nucleus at low temperatures and high relative humidity with respect to ice, because emissions are much higher than those of biological material and dust particles, soot is still able to play an important role in cloud formation and climate (Chou *et al.* 2013).

5 Organic aerosols (glassy solids)

Aqueous organic aerosols are prevalent in the atmosphere, and under cirrus conditions, or at altitudes of 7-16 km, have been found to become glassy (Murray, 2008; Quante, 2004; Zobrist *et al.*, 2008). For instance, through the use of a laser ionization mass spectrometer (PALMS) in conjunction with time of flight mass spectrometry, Murphy *et al.*, (1998) found that upper tropospheric particles containing sulfate are frequently internally mixed with organic constituents. In addition, field measurements conducted in urban and remote areas using an AMS have indicated that atmospheric aerosols contain a substantial fraction of organic material (18-70%), especially in the more anthropogenically impacted northern hemisphere (Zhang *et al.*, 2007). Using Aerodyne AMS to take quantitative measurements of the size and chemical composition of non-refractory submicron aerosol particles, Alfarra *et al.* (2004) found that the total dry fine particle mass in the atmosphere typically consists of 10-70% low-volatility organic compounds.

Aqueous solution droplets containing oxygenated organic compounds have been discovered to become glassy in the troposphere (Murray, 2008). The constituent molecules of glassy solids lack the long range order that is characteristic of crystals. Such amorphous solids form when a liquid is cooled to a temperature at which the molecules stop diffusing, and its viscosity increases correspondingly, resulting in them being held in a "liquid-like" amorphous state (Koop *et al.*, 2011; Murray, 2008).

Ice nucleation on glassy organic aerosols has only become of great interest relatively recently. As a result, the available quantitative information does not yet provide a strong conclusion regarding their influence on ice nucleation. For instance, Zobrist *et al.* (2006) found that oxalic acid behaves as heterogeneous immersion ice nuclei by initiating freezing at temperatures 1.8-2.0 ^{o}C warmer than that of pure water. Murray (2008) concluded from studies regarding citric acid droplets that glassy organics inhibit ice nucleation even when conditions are sufficiently supersaturated for homogeneous nucleation. These results are as expected due to the amorphous character of glassy organics, which would reduce both their crystallographic match with ice and therefore their ability to function as good ice nuclei.

In contrast, Baustian *et al.* (2010) found that while ammonium sulfate initiated depositional ice nucleation at an average ice saturation ratio of 1.10 ± 0.07 between -38 ^{o}C and -59 ^{o}C, glutaric acid required an ice saturation ratio that increased from 1.2 at -38 ^{o}C to 1.6 at -59 ^{o}C. Shilling *et al.* (2006) found that the critical saturation ratio for depositional ice nucleation on ammonium sulfate and maleic acid ranged from 1.04 ± 0.05 at -33 ^{o}C to 1.42 ± 0.04 at -83 ^{o}C. Due to these conflicting results it is evident that more quantitative characterization of the ice nucleation properties of glassy organic is required in order to better understand their role in the atmosphere. Recently, Ladino *et al.* (2014) found that secondary organic aerosol particles are able to nucleate ice via deposition nucleation if the particle viscosity is increased by pre-cooling.

6 Conclusions

Shown below in Fig. 3 is a compilation of onset ice nucleation temperatures and saturation ratios of a variety of particles that have been found to be atmospherically relevant in recent studies (Hoose and Möhler, 2012). This figure demonstrates that while many studies have been conducted on mineral dusts, bio-aerosols, soot, and organic aerosols, there is not necessarily good agreement between or within the four modes of ice nucleation (deposition nucleation, condensation freezing, immersion freezing, and contact freezing). As a result there are still gaps in the understanding of ice nucleation mechanisms that must be filled before any final conclusions can be drawn. These discrepancies directly impact the developed parameterizations which are commonly used in climate models to infer the future climate. Therefore, the climate predictions of the upcoming decades and the impact of global warming possess significant uncertainties which must be reduced without delay.

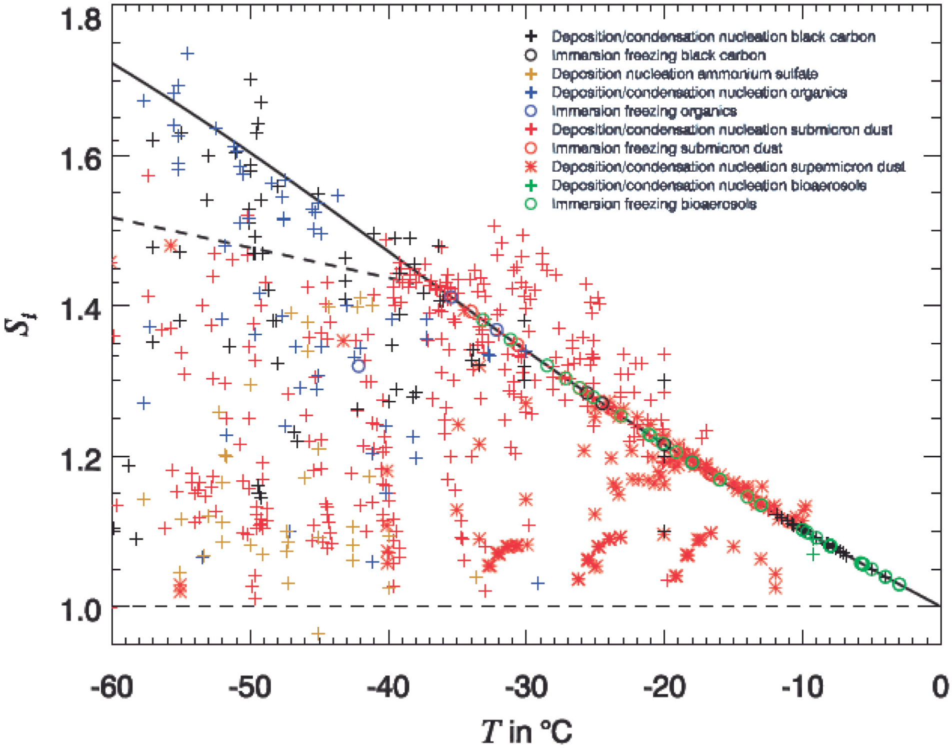

Figure 3. *Compiled ice nucleation onset temperatures and saturation ratios,* S_i *(Hoose and Mohler 2012).*

The tropical countries can locally and globally contribute to ice cloud formation due to the following reasons: There are several arid areas within the tropics (e.g. Atacama Desert (Chile), Saharan Desert (North and Central Africa), Sechura Desert (Peru) and Simpson Desert (Australia)) which emit mineral dust particles into the atmosphere. As shown above, those mineral dust particles could efficiently nucleate ice clouds at low relative humiditiy.

Although several global climate models indicate that the influence of bio-aerosols is very small on a global scale, they may have a substantial effect in the tropics due to the large amount of land that is covered by forests which emit many biological aerosol particles.

Because the tropics are largely associated with marine environments (e.g. Atlantic, Pacific and Indian Oceans), marine aerosols are present in the tropical atmosphere. Those particles could locally or globally influence both the radiative balance of the planet and the hydrological cycle. However, because our understanding of them is poor, their impact is not well known.

For each of these particle sources, the potential for lofting to high altitudes where cirrus clouds and mixed phase clouds may form is considerable given the convective activity that occurs in the tropics.

Similarly, tropical countries largely contribute of the total amount of soot particles that are injected into the atmosphere due the poor control of gas and particulate matter emissions from industries, transportation, and biomass burning. As a result, if soot particles are able to nucleate ice under certain conditions, the tropics could be a very important source of IN and therefore of ice clouds.

Tropical countries and the international community should focus their attention on the tropics with the aim of characterizing the aerosol particles that are emitted by those countries with the goal of inferring the role that they play within the local and global climate. As shown above, the tropics have the ability to emit a large number of aerosol particles which have been identified as efficient ice nuclei. Therefore, they can have a significant and global influence on the Earth through their impact on its radiative balance and hydrological cycle.

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