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Discrepancy between Ice Particles and Ice Nuclei in Mixed Clouds: Critical Aspects

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Abstract

Measurements of ice crystal concentrations in mixed clouds tend to exceed ice nucleus concentrations measured in nearby clear air. This discrepancy is a source of uncertainty in climate change projections as the radiative properties of mixed phase clouds are largely determined by their liquid and ice water content. The ice enhancement process can sometimes depend on secondary ice production, which can occur through ice crystal fracture during sublimation, cloud drop shattering during freezing or following collision with ice particles. However, the discrepancy is observed even in mixed clouds where only primary ice nucleation processes occur. Several hypotheses have been suggested for the observed discrepancies. One factor could be the existence in clouds of pockets of high vapor supersaturation formed by droplet freezing or removal of small droplets by collision with larger droplets, associated with the fact that ice crystal concentration increases with water supersaturation. However, ice crystal concentrations are usually measured at near water saturation. Additional factors could be drop freezing during evaporation and activation of droplet evaporation residues. Here we suggest that a major factor could be underestimation of the contact freezing mode as it is not measured in experimental campaigns and seldom considered in nucleation models. Laboratory experiments give only incomplete answers to the important questions concerning the contact freezing mode, e.g. what fraction of the aerosol particles that come into contact with the droplet surface results in a freezing event and what is the influence of particle type and size, air temperature and relative humidity. As supercooled droplets grow or evaporate in mixed clouds, phoretic forces should play an important role in the collision efficiency between aerosol and droplets, and consequently in contact freezing. A further question is the possibility that aerosol, usually not active in deposition or condensation/immersion freezing, can trigger ice nucleation by colliding with supercooled droplets.

Keywords

Ice Crystal, Ice Nuclei Particles, Ice Nucleation Process, Thermophoresis, Diffusiophoresis

1. Introduction

Microphysical processes in clouds are important for cloud evolution, precipitation and the atmospheric environment. Incomplete knowledge of the ice nucleation processes is one of the main obstacles to successful implementation of cloud microphysical models. Ice can form through primary processes (nucleation from the liquid or water vapor phases), homogeneously or heterogeneously triggered by aerosol called ice nuclei particles (INP), and secondary processes that produce ice crystals in the presence of pre-existing ice without requiring the action of ice nucleating particles.

Supercooled droplets are quite common in clouds. Pure micrometric water drops freeze spontaneously (homogeneous process) if the air temperature is below about −35˚C, while haze droplets freeze at lower temperatures. In the presence of INP, ice formation initiates at a higher temperature than homogeneous nucleation, in a variety of ways: deposition, condensation freezing, immersion freezing and contact freezing. Therefore, in cold clouds at temperatures below −35˚C, the dominant ice nucleation mode should be homogeneous freezing, while at higher temperatures ice nucleation should take place heterogeneously. Substances have different freezing efficiency depending on the nucleation mode, indicating that the freezing mechanism differs for the different modes, further hindering our understanding of ice formation.

Here we discuss one of the puzzling aspects observed in mixed clouds, where ice particle number concentrations are sometimes much higher than those expected from primary ice nucleating particle concentration measured in nearby clear air. This discrepancy is a source of uncertainty in climate change projections as the radiative properties of mixed phase clouds are largely determined by their liquid and ice water content [1].

2. Ice Enhancement Processes

Although the problem of ice enhancement has arisen much interest and debate in cloud physics and several possible explanations for the phenomenon have been proposed, uncertainties remain on which mechanisms are responsible for the high particle concentrations observed in the wide range of cloudy conditions. To try to understand the relationship between ice particles and INP concentrations we will consider previously published papers related to events in which the relatively high ice crystal concentration is not ascribable to secondary ice generation.

2.1. Primary Ice Production

Ice crystal number concentrations in mixed clouds tend to exceed ice nucleus

concentrations in nearby clear air, as determined from standard ice nucleus measuring techniques [2]-[7]. This is particularly evident in clouds with a cloud-top temperature warmer than about −12˚C [8] [9] [10] [11]. Several hypotheses have been suggested for the observed discrepancies.

2.1.1. Water Vapour Supersaturation

For experimental campaigns not involving artefacts (e.g. ice crystals shattering on the inlet probes during aircraft campaigns) and in secondary ice production, one factor reported in published papers for enhanced nucleation mechanism is the possible existence in clouds of pockets of high vapor supersaturation [3] [12]. It is known that ice crystal concentration increases with water supersaturation Sw. High supersaturation can be related to:

1) Droplet freezing. When ice is nucleated in a supercooled drop, the drop temperature quickly rises to 0˚C and the drop is a source of heat and water vapor. Large values of Sw can occur around the drop (>10%), and this may lead enhanced primary ice nucleation [13].

2) Removal of small droplets by collisions with larger droplets during a rising parcel of air [4]. Within this region of high saturation ratio with respect to ice and water (up to 15%) aerosol particles can be activated as cloud condensation nuclei or even INP, and rapid ice nucleation may occur to produce ice crystal concentrations greatly in excess of those measured by ice nucleus counters, which generally operate close to water saturation. Further processes could help clarify the discrepancy between ice particle and ice nuclei concentrations.

2.1.2. Drop Freezing during Evaporation

By considering pure water droplets, Santachiara *et al*. [14] showed that small ice crystals growing in the presence of supercooled droplets (unstable system) trigger the freezing of fast evaporating droplets (Wegener-Bergeron-Findeisen process) even at much higher temperatures (range −12˚C ÷ −14˚C) than those considered for homogeneous nucleation in stationary conditions. These results support field campaigns, where an increase in ice crystal concentration by factors of 100 - 1000 in less than 10 minutes was observed in the presence of a thin layer of supercooled water drops and a few ice crystals at the top of stratiform, wave and convective clouds (temperatures between −6˚C and −18˚C). Rangno and Hobbs [15], studying convective clouds (Marshall Island, in the western Pacific ocean), observed that extremely high concentrations of ice particles (often > 500 l^{-1}) formed very rapidly at temperatures between −4˚C and −10˚C and were initiated by the freezing of individual drops. Ansmann *et al*. [16], in tropical liquid and mixed-phase altocumulus cloud, found that the liquid phase forms first, before ice crystals nucleate in the altocumulus layers, and concluded that ice nucleation starts when evaporating supercooled droplets freeze due to entrainment of dry air. These events could be explained following the previously cited laboratory experiments.

Growth of initial ice crystals can determine homogeneous or heterogeneous freezing (e.g. inside contact) of the droplets as fast evaporation determines a

high cooling rate [6] [17] [18] [19] [20]. The laboratory results of Santachiara *et al*. [14] could even help explain the so-called cloud "seeder-feeder" mechanism. In this process, ice crystals from cirrus or higher altocumulus layers fall into lower liquid layers and trigger significant ice production at temperatures that are usually too high for the initiation of heterogeneous ice formation. The "seeder-feeder" mechanism can happen even in mixed cloud where "seeder" and "feeder" zones exist. If ice crystals ("seed crystal") fall through the supercooled liquid water cloud into the lower layers of the cloud ("feeder zone"), they grow due evaporation of droplets or riming, and can even determine their freezing.

2.1.3. Activation of Droplet Evaporation Residues

Rosinski and Morgan [21] found that drop evaporation leaves a residual (*i.e*. a particle soluble, insoluble or mixed) that can subsequently act as ice nuclei if water vapor is supersaturated with respect to ice. Ice formation in the Arctic mixed-phase clouds may be explained by the activation of droplet evaporation residuals, and drop freezing during evaporation [7] [22].

2.1.4. Underestimation of the Contact Freezing Mode

Contact freezing is defined as the process in which freezing of a supercooled droplet results from its collision with an aerosol particle. It is known from laboratory experiments that particles acting as contact nuclei nucleate at higher temperatures than particles embedded in non-evaporating drops, and that contact freezing can occur in droplets from the inside-out as well as from the outside-in [23] [24] [25] [26].

In the laboratory, contact freezing can be measured with several devices (cold plate technique, thermal diffusion chamber, electrodynamic balance, wind tunnel, etc), but no instrument has yet been devised to measure all nucleation modes and to isolate contact nucleation in clouds. Laboratory experiments [25] [27] [28] [29] give only incomplete answers to the important questions concerning the contact freezing mode, *i.e*. are the same ice active sites active in immersion and in the contact freezing mode? What fraction of the aerosol particles that come into contact with the droplet surface result in a freezing event? And what is the influence of particle type and size, air temperature and relative humidity? A summary of the key questions on the impact of contact freezing on cloud glaciation can be found in Ladino *et al*. [30].

Results reported by Durant and Shaw [24] reveal very similar contact freezing temperatures for volcanic ash, glass-rich volcanic ash and soda glass particles. Fornea *et al*. [25] found that the most effective contact freezing INP was Pahokeee Peat soil (−10.5˚C), followed by volcanic ash (−11.2˚C), and finally soot (−25.6˚C). Contact freezing activity was found to increase with increasing particle size [28] [31] [32] [33]. Generally speaking, contact nuclei appear to be less dependent than immersion freezing on temperature, aerosol type and droplet size. Several experimental campaigns need to consider contact nucleation in order to account for ice crystal concentration [5] [6] [7] [11] [34].

For experimental evaluation of contact-induced nucleation, it is necessary to

know the collision efficiency between droplets and aerosol, and select which particles can initiate ice formation (contact nuclei). Liu *et al*. [35], Lohmann *et al*. [36] assumed that all dust aerosols can act as contact nuclei. Phillips *et al*. [37] proposed a contact freezing parameterization by assuming that each ice nucleus particle can nucleate ice at a freezing temperature that is 4.5 higher than the freezing temperature associated with immersion or condensation freezing. Gettelman *et al*. [38] assumed that only coarse dust aerosols can act as contact ice nuclei.

Concerning the interaction with droplets, aerosol particles can be scavenged from the air by liquid drops through Brownian diffusion, interception, inertial impaction, electroscavenging, thermophoresis and diffusiophoresis (phoretic forces). Experimentally, the collision efficiency can be determined by exposing a droplet to the aerosol flow in the absence of freezing, and then evaporating the droplet and counting the residual aerosol [29] [32]. The freezing efficiency, defined as the freezing probability per droplet-particle collision, can then be derived. Freezing efficiencies obtained with an electrodynamic balance for illite and kaolinite were measured by Hoffmann *et al*. [32] [33], and Svensson *et al*. [27] in high, intermediate, and dry conditions. Svensson *et al*. [27] observed a higher freezing threshold at higher humidity with respect to dry conditions. Niehaus *et al*. [29] obtained similar freezing efficiency for Arizona test dust, feldspar and rhyolitic ash. For the mineral, an efficiency of about 10^{-5}, increasing by decreasing temperature, was measured at −15˚C. Snomax showed a freezing efficiency of 10^{-3} at −5˚C. These results were obtained in the absence of phoretic forces.

However, it is important to note that droplet growth and evaporation in clouds occur frequently, along with concomitant thermo- and diffusiophoretic forces, which act in opposite ways. Thermophoresis pushes particles toward lower temperatures (*i.e*. towards an evaporating droplet), whereas diffusiophoresis with Stefan flow moves the aerosol in the opposite direction [39]. Therefore, phoretic forces influence collision efficiency and consequently the contact freezing efficiency.

There is disagreement between laboratory experiments and theoretical models concerning the prevalence of thermophoresis or diffusiophoresis when droplet growth or evaporation occurs, thereby increasing the uncertainty in the parameterization model. The impact of phoretic forces is sometimes neglected [35], or only the thermophoretic process is considered, neglecting diffusiophoresis [40] [41], or a prevalence of thermophoresis over diffusiophoresis is assumed. These statements are questionable. Published experiments show a prevalence of Stefan flow with respect to thermophoresis in case of evaporating droplets [42] [43] [44], in disagreement with theoretical papers [45] [46] [47].

Several laboratory experiments show a higher collection efficiency in the Greenfield gap at high with respect to lower Relative Humidity (RH), indicating a prevalence of diffusiophoresis with Stefan flow over thermophoresis. Ladino *et al*. [48] in experiments performed at RH = 90% (droplet evaporating with d =

25.6 μm), showed (Figure 7 of the cited paper) a lower collection efficiency in the Greenfield region with respect to Grower *et al*.'s model (1977) [46] where Brownian motion and phoretic forces are included, based on a prevalence of thermophoretic force. Ardon-Dreyer [49] found higher collection efficiency in the Greenfield region at high RH (88%) with respect to lower RH (11%) at room temperature (22.5˚C) and a droplet diameter of 43 μm. Even Svensson *et al*.'s results [27] should confirm a prevalence of hydrodynamic Stefan flow.

Additional processes can interfere in the contact mode. An important question is whether the homogeneous nucleation process in supercooled water droplets can occur not only in the interior volume of the droplet, but even at or close to its surface [50]. Therefore, contact nuclei could favor surface nucleation. An additional factor that could confirm an underestimation of the contact freezing mode is the possibility that aerosol, usually not active in deposition or condensation/immersion freezing, can trigger nucleation by colliding with supercooled droplets. With respect to this question, an important result was obtained by Niehaus *et al*. [51] who found that the impact of water soluble salt particles initiated freezing in experiments using water droplets at supercooling temperatures of 9˚C to 16˚C. These results show that contact freezing nuclei can even be particles not active in deposition or immersion freezing. Figure 1 shows a schematic view of the water nucleation processes suggested to clarify the discrepancy between INP and ice particle concentrations in mixed clouds.

2.2. Secondary Ice Production

In several cases, the discrepancy between ice crystals and INP can be explained by considering secondary ice formation processes. Some published papers report that the high concentration of ice is initiated by primary processes such as freezing of individual drops, some of which can fragment upon freezing, accompanied by ice splinter production during riming. Secondary ice formation

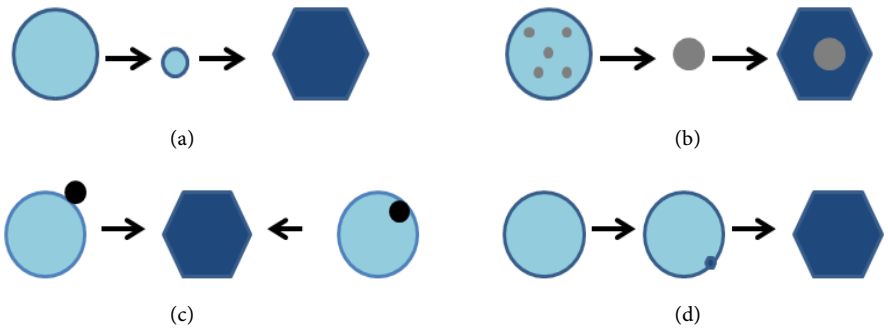

Figure 1. A schematic view of the water nucleation processes suggested to clarify the discrepancy between INP and ice particle concentrations in mixed clouds. (a) Evaporation freezing; (b) Activation of droplet evaporation residuals; (c) Contact nucleation: contact nucleation can occur in droplets from inside-out, as well as from outside-in; (d) A homogeneous ice nucleation process (pure water or solution droplets) can start on the surface instead of inside the droplets. Symbols: circles (light-blue), water droplets; small circle (grey), residual of droplet evaporation; small circle (black), aerosol particle; hexagon, ice crystal.

processes can occur through:

1) Fracture of ice crystals during sublimation [52] [53] or following collision of preexisting ice crystals [54];

2) Shattering of large isolated cloud drops during freezing in free fall [55] [56];

3) Hallett-Mossop process, *i.e.* fragmentation of freezing droplets following their collision with ice particles, e.g. graupels, snowflakes [57] [58] [59].

There is a consensus that the Hallett-Mossop process can occur within a temperature range of approximately −3˚C to −8˚C, requires a 0.2 - 5 $m \cdot s^{-1}$ impaction speed and the presence of cloud drops both smaller than 13 μm and larger than 24 μm. One or more of these processes can be present in the examined clouds [60]. Modeling studies mainly consider only the Hallett-Mossop rime splintering process. A review of secondary ice production can be found in Field *et al*. [61].

3. Conclusion

To help solve the thorny problem of the discrepancy between ice particle and ice nuclei concentrations, laboratory experiments should be extended to shed more light on the processes that control ice initiation, in addition to classical nucleation modes. Future studies should consider the possible activation of droplet evaporation residues, droplet evaporation freezing of pure or solution droplets, the presence of high supersaturation in clouds, and the mechanisms underlying contact nucleation. This should determine the impact of phoretic forces on the collision between aerosol and droplets during growth and evaporation. Additional investigations should address the possibility that aerosol, usually not active in the deposition or condensation/immersion freezing, can trigger nucleation by colliding with supercooled droplets.

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