Scientific Research Publishing

Atmospheric and Climate Sciences, 2023, 13, 466-477
https://www.scirp.org/journal/acs
ISSN Online: 2160-0422
ISSN Print: 2160-0414

A Review of Thermo- and Diffusio-Phoresis in the Atmospheric Aerosol Scavenging Process. Part 2: Ice Crystal and Snow Scavenging

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How to cite this paper: Santachiara, G., Prodi, F., Belosi, F. and Nicosia, A. (2023) A Review of Thermo- and Diffusio-Phoresis in the Atmospheric Aerosol Scavenging Process. Part 2: Ice Crystal and Snow Scavenging. *Atmospheric and Climate Sciences*, **13**, 466-477.
https://doi.org/10.4236/acs.2023.134026

Received: June 16, 2023
Accepted: October 5, 2023
Published: October 8, 2023

Abstract

The role of phoretic forces in the identification of particles acting as ice nuclei in mixed phase cloud is discussed. A method used to identify the effective ice nucleating particles is to sample ice crystals, which are afterwards sublimated, and to examine the particles remaining after evaporation. The procedure takes into account only crystal with a maximum diameter of 20 μm, by assuming that small crystals do not scavenge aerosol during growth, and therefore that crystals contain only the effective nucleating particles. This assumption is questionable, however, as experiments have shown that even small ice crystals can scavenge aerosol. Another approach has been to compare the number and elemental composition of residual particles in small ice crystals and of aerosol near the cloud. By considering as example soot and black carbon aerosol, contradictory conclusions on their importance in the processes of ice nucleation have been reported in the literature. We suggest that, in addition to physico-chemical properties of soot/carbon aerosol particles, even the microphysical and environmental parameters involved in the transition of aerosol from gas phase to ice crystals in cloud should be considered. The contribution of phoretic forces should also be considered. After initial growth ice crystals can continue to grow by water vapour diffusion. Laboratory experiments confirm the contribution of diffusiophoresis with Stefan flow in the scavenging by snow crystals up to 3 mm in diameter. The particle scavenging efficiency of snow crystals is related to crystalline shape and depends on air relative humidity and temperature.

Keywords

Ice Crystals, Snow Crystals, Ice Nucleating Particles, Aerosol Scavenging,

Phoretic Forces

1. Introduction

Atmospheric clouds and precipitations (rain, snow, hail or fog) play a fundamental role in the removal of atmospheric aerosol particles. Wet removal of aerosol particles from the atmosphere includes in-cloud scavenging (wash-out) and below-cloud scavenging (rain-out). Clouds can be classified as liquid, mixed (when ice particles coexist with supercooled water droplets) or iced, if the ice phase (as in cirrus clouds) is prevalent.

The formation of liquid clouds requires the presence of aerosol particles called cloud condensation nuclei (CCN), while the formation of ice clouds can happen homogeneously or heterogeneously, when aerosol particles, called ice nucleating particles (INPs), promote the ice phase. INPs may represent about 1 in 10^5 of all aerosol particles in the free troposphere.

In-cloud aerosol scavenging is due to nucleation and impaction, which include Brownian motion, interception, inertial impaction, thermophoresis, diffusiophoresis, electrostatic attraction and air flow turbulence. Brownian diffusion dominates scavenging of particles with radius of less than 0.01 μm and inertial impaction of particles with radius over 1 μm. Phoretic forces are effective in the size range 0.01 ÷ 1 μm [1].

In a previous paper, Santachiara *et al.* [2] examined in-cloud and below-cloud atmospheric scavenging due to falling drops. Several experimental studies observed in droplet growing an increase of aerosol scavenging, due to diffusio-phoresis with Stefan flow [3] [4]. These results were later confirmed by Feng *et al.* [5]. Prodi *et al.* [6] also confirmed these results during an experimental campaign performed at the Drop Tower Facility (Bremen) in microgravity conditions, when they observed that an evaporating droplet pushes the particles away. In contrast, during droplet growth due to condensing water vapour, aerosol was pushed towards the droplet surface.

As to ice formation in clouds, homogeneous nucleation requires temperature lower than −38˚C. In the presence of INPs, primary ice formation initiates at a higher temperature than homogeneous nucleation and in four freezing modes: deposition nucleation (water vapour is transformed into ice on an INP surface), condensation freezing (ice formation during liquid condensation), immersion freezing (freezing by a particle immersed in a liquid drop), and contact freezing (freezing when an aerosol particle collides with a liquid droplet).

More recently it has also been shown that in porous particles, water condensation in subsaturated conditions followed by ice nucleation could be an alternative to deposition nucleation [7].

Ice nucleation processes have been widely researched both in the laboratory and the field. An overview on INP particles was presented by Kanji *et al.* [8]. Experimental measurements have evidenced the most relevant natural sources of

INPs (*i.e.* deserts, volcanic eruptions) and important anthropogenic sources (*i.e.* agricultural activity, deforestation, biomass burning). Recent studies also show that sea spray aerosols may be an important source of INPs in remote marine regions [9].

However, equating the results of laboratory measurement performed under controlled conditions and using specific aerosol particles with the atmosphere is problematic. For instance, while ice active surface site density (n_s) is measured in the laboratory for a single type of aerosol, in air masses n_s is calculated by assuming that all particles are INPs even if the majority of the particles will not activated. Therefore, the n_s of the aerosol population will be lower than the value obtained in a laboratory test [10] [11].

The present paper looks at issues regarding the identification of INPs in the ice crystals and the possible role of phoretic forces in the ice crystal scavenging. A brief overview of the complete growth cycle of the ice phase and related scavenging modality are also presented.

2. Identification of Ice Nucleating Particles in the Atmosphere

The identification and detailed characterization of particles that act as IN in mixed phase clouds is important for a better understanding of the ice-forming processes. Information on the composition of ice nuclei in the real atmosphere can be obtained by sampling cloud ice crystals and examining the particles they contain.

In the past, ground-based examinations directly identified particles present in ice crystals off-line with electron microscope techniques [12]. Kumai and Francis [13] studied sampled snow crystals for nuclei identification at an altitude of 2000 m, about 320 km east of Thule on the Greenland Ice Cap, and observed "innumerable minute particles" in the remainder of the crystals (prevalently hexagonal plate and dendritic crystals). This showed that growing crystals scavenge aerosol particles. As a result, the authors suggested that a vapour pressure gradient between the surface of snow crystals and their environment during growth, *i.e.* diffusiophoresis, caused atmospheric particles to be drawn into the snow crystals.

A subsequent approach was to activate aerosol directly in the atmosphere (*e.g.* with a continuous flow ice-thermal diffusion chamber), sampling ice crystals via impaction onto substrates for off-line microscopy analysis [14]. In the early 1990s efforts were made to understand the nature of INPs using a counter-flow virtual impactor (CVI) to separate ice crystals from other solid/liquid components (interstitial particles, supercooled droplets, etc.) in mixed-phase clouds [15]. The ice crystals were afterwards sublimated and the residual particles, *i.e.* particles remaining after evaporation of ice, collected for off-line analysis.

More recently real time aerosol mass spectrometry techniques, including single particle analysis by laser mass spectrometry (PALMS) and aerosol mass spectrometry (AMS) have been used for aircraft-based measurements [16] [17] [18].

However, despite the complex procedures adopted, whether INPs, were unambiguously identified remains a contentious issue. During crystal growth, for example, additional particles could be transferred to ice crystals. As a result, particles left after ice evaporation of crystals may not be identical to INPs.

Another method used to identify ice nuclei was to compare the number or mass and chemical compositions of ice crystal residuals and the background aerosol [19] [20]. High enrichment (*i.e.*, the ratio between the percentage in number or mass of the particle type or group of particles found in ice crystal residuals compared with background aerosol) was considered a valid criterion to identify the ice nuclei particles. We will address to these modalities separately.

3. Particle Scavenging by Small Ice Crystals

One approach employed to identify the effective ice nucleating particles is to sample only crystals with a maximum diameter of 20 μm, on the questionable assumption that ice crystals up to this size do not present scavenging. An ice crystal should grow by water vapour diffusion to 20 μm in a water saturated environment in about 20 s [21]. The transition from diffusional growth to riming is predicted to take place for ice crystals larger than 50 μm [22].

During an experimental campaign carried out at Dome C (Antarctica) in the period 21 February-6 August 2012, falling ice crystals were sampled during clear-sky and cloud precipitation events [23]. Replicated crystals (hexagonal plates, pyramids, solid and hollow columns, needles) were examined at Scanning Electron Mycroscopy (**Figure 1**). Riming was not observed, *i.e.* the only process of crystal growth was water vapour diffusion. As scavenged aerosol was observed even in small crystals—a finding that could not be explained by Brownian motion, inertial impaction and interception—it was concluded that diffusiophoretic forces may play a role in the collection of aerosol. During diffusive crystal growth, a flow, known as Stefan's flow, is present near the surface of the growing crystal, and the motion of the particulate is the same as that of water vapour flux [24].

Santachiara *et al.* [25] have showed even in laboratory experiments that ice crystals with diameters of 10 - 30 μm scavenged insoluble aerosol in the early stage of growth (**Figure 2**), and moreover, that scavenging due to Brownian diffusion, impaction, and interception could not account for the scavenged aerosol observed. The Authors suggested that the presence of aerosol in the small crystals could be explained by diffusiophoretic force due to water vapour gradient between droplets and ice crystals. However, the key finding in both the conducted experiments was that even crystal of about 20 μm scavenged aerosol particles during growth.

Other papers in the literature reporting on experimental campaigns also discuss possible particle scavenging in crystals with diameter of about 20 μm. Soluble compounds, usually regarded as inefficient INP, were measured in ice particle residuals in cold and/or mixed clouds at Jungfraujoch station [18] [20] [26]

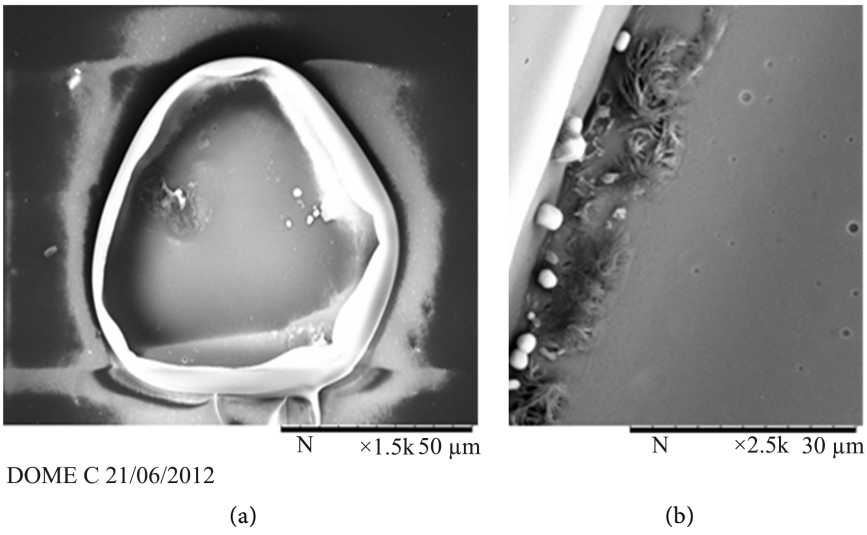

Figure 1. Replicas of ice crystals sampled on 21 June 2012: (a) Hexagonal plate; (b) Sodium chloride aerosol on the edge of an ice crystal.

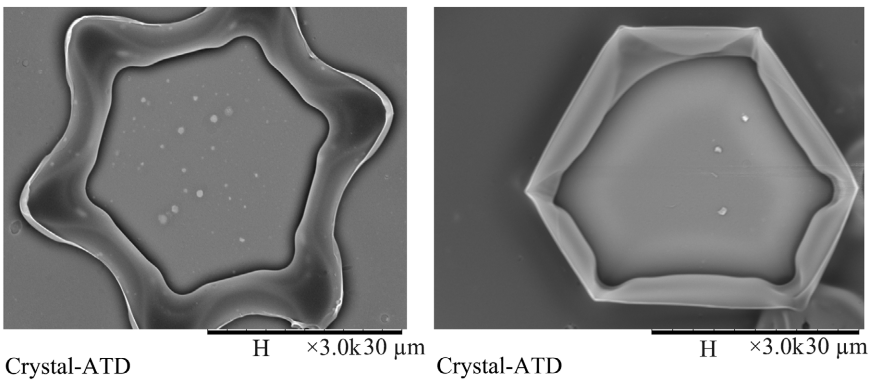

Figure 2. Ice crystal replicas showing scavenged aerosol particles.

Even if sampled ice crystal diameter was lower than 20 µm, these authors recognized the possibility of salt/soluble compounds scavenging process.

When sampling small ice crystals in a joint field campaign (January-February 2013, Jungfraujoch), Worringen *et al.* [27] observed considerable amounts of inexplicable soluble material (*i.e.* sea-salt, sulphates), which they suggested might be instrumental contamination artifacts. In our view, however, any "artifact" should be produced by all the three different techniques used to separate INPs as well as ice particle residuals from interstitial aerosol, a possibility that appear unlikely. More probably, there was an aerosol removal process after nucleation, even if the crystals were smaller than 20 µm.

4. Particle Residual in the Ice Crystals Compared with Background Aerosol

A further mean of identifying ice nucleating particles in ice crystals is to compare the chemical composition of the residual particles in small ice crystals with those near the cloud. The transfer of aerosol in mixed clouds from air to ice

phase involves several steps, which depend on the mode of ice nucleation. The complexity of the process to effectively identify INPs is best illustrated by a brief review of the results published on the ice nucleation efficiency of soot and black carbon (BC) particles. Major anthropogenic pollutants, soot and BC particles are mainly composed of highly agglomerated carbon spherules generated in the incomplete combustion of fossil fuels and biomass [28].

Of note is the fact that laboratory and field measurements show contradictory results on the effectiveness of soot and BC particles to promote the formation of the ice phase in the atmosphere. Laboratory experiments conducted by Vergara-Temprado *et al.* [28], Friedman *et al.* [29], Schill *et al.* [30], Mahrt *et al.* [31] and Kanji *et al.* [32], in water-saturated conditions, imitating mixed-phase clouds dominated by condensation and immersion freezing modes, detected low BC efficiency at T > 240 K and with a variety of soot types. The Authors concluded that if the carbonaceous particles in the atmosphere behave like those used in the aforementioned laboratory studies, BC is unlikely to play a substantial atmospheric role as an INP via the immersion mode in mixed-phase clouds.

Field research comparing residual aerosol in ice crystals and bulk aerosol performed by Kamphus *et al.* [26], Pratt *et al.* [33], Kupiszewski *et al.* [34], Hammer *et al.* [35] and Schill *et al.* [36] showed low ice nucleation efficiency of BC particles. However, other field studies have shown soot to have higher levels of ice nucleation activity [19] [37] [38] [39]. Phillips *et al.* [40] suggested that black carbon is a major INP in clouds influenced by biomass-burning-derived particles.

Nor do the conclusion of field measurements performed during the same campaign agree. For example, a strong correlation between number concentrations of soot and biomass-burning-derived aerosols and ice concentrations was observed in a mixed-phase orographic wave cloud at high altitude over Wyoming during the Cloud experiment-Layer Clouds (ICE-L), suggesting that BC might nucleate ice [41]. However, in the same campaign Pratt *et al.* [33] found that the percentage of BC in the ice crystal residuals was low. Despite conflicting results, current global models often identify BC as the second-most abundant INP type after mineral dust.

The contradictory conclusions as to the importance of soot and BC in the ice nucleation atmospheric process can depend on the great variability of the physico-chemical properties of soot/carbon aerosol particles, the variety of sources and aging processes. Freshly emitted soot particles, for example, are mostly hydrophobic, *i.e.* they barely act as CCNs. Atmospheric aging particles often results in BC becoming coated with secondary aerosol material (such as inorganic ions). This aged BC exhibits greater hygroscopicity, with the result that the transfer of BC into cloud droplets via nucleation scavenging is enhanced [42].

5. Discussion

As well as the chemico-physical properties of the aerosol, it is also important to consider the microphysical processes involved in the transfer of interstitial

aerosol to ice phase. Immersion freezing nucleation in mixed clouds follows several steps: activation of aerosol as droplet, droplet freezing, followed by the growth of ice crystals. These steps include water vapour diffusion and aerosol scavenging processes.

The residual of ice crystals in immersion freezing mode therefore consists of particles that activate droplets and ice (CCN, INPs) and any scavenged particles. A subset of CCN may also be effective INPs. In both condensation and deposition freezing, the residual consists of particles on which the water vapour condense and/or freeze and of aerosol scavenged during crystal growth.

Air temperature and relative humidity influence the scavenging of aerosol particles as affect the rate of crystal growth. In mixed-clouds, ice crystals grow when the vapour pressure in air (e_{air}) is lower than vapour pressure of water (e_w) and higher than the ice water vapour e_i (Wegener-Bergeron-Findeisen process). Any variation of growth rate implies a variation of both water vapour flux and the aerosol particles carried by the vapour. The WBF process has a dual role in clouds, *i.e.*, on the one hand, it favors aerosol scavenging by the crystal, and on the other, it contributes to the release of the previously activated particles back into the interstitial phase [43].

It follows that atmospheric thermodynamic conditions (*i.e.* air temperature, pressure and relative humidity), which influence the nucleation processes, particle concentration scavenged by ice crystals, and therefore, evaluation of the nucleation efficiency of aerosol particles, should be considered in field campaign intercomparison. Failure to consider these parameters could contribute to explain the contradictory results obtained in the above-mentioned studies. Scavenging processes also depend on liquid water content (LWC) and ice water content (IWC) in clouds [44] [45].

To evidence that ice crystal ≤ 20 μm can scavenge aerosol particles, we will consider Jungfraujoch site, where mixed phase clouds dominate the cloud microphysics and WBF is active [46] [47]. Assuming a total number aerosol concentration of about 1000 cm^{-3} [46] the theoretically collection efficiency of small ice crystal due to Brownian diffusion, interception and inertial impaction, based the Fuchs formula [48] and Park *et al.* [49] was seen to be negligible for crystals ≤ 20 μm.

Collection efficiency might be increased as a result of diffusiophoresis, due to e_{air} higher than e_i and lower than e_w. In fact, assuming a number concentration of droplets and ice crystals of about 400 cm^{-3} and 1 cm^{-3}, respectively [47], the resulting high ratio between droplet/crystal number causes high vapour flux toward individual crystals and consequently of interstitial aerosol particles [24].

After initial growth, ice crystal can continue to grow by water vapour diffusion. Laboratory experiments were performed by Vittori and Prodi [50] considering ice crystals growing surrounded by supercooled droplets in the presence of fluorescent ZnS particles, which were captured on crystal branch tips. A dust free space was observed near evaporating droplets. Further experiments confirmed these results with sodium chloride and carnauba wax [51].

Oraltay and Hallett [52] investigated the scavenging efficiency of particles by ice crystals growing in supercooled clouds. When growing crystal reached about 3 mm, polydispersed soot was injected and aerosol was collected efficiently due to diffusiophoresis.

In field processes further ice growth modes are present. Ice crystals can aggregate to form snowflakes or grow by inertial scavenging of cloud droplets, which subsequently freeze (riming).

The scavenging of aerosol by ice crystal and snow is important in remote polar regions where wet deposition due to ice phase is prevalent. In Antarctica pristine crystals (plates, pyramids, columns, bullets) are prevalent in the ice phase [53] and as crystal growth in such remote area is mainly due to water vapour diffusion, phoretic forces could be crucial in aerosol removal. The particle scavenging efficiency of snow crystals is relayed to crystalline shape with needles and columns less effective than stellar plates, dendrites and snowflakes. The collection efficiency depends on air relative humidity and temperature, indicating a contribution of phoretic forces [54].

6. Conclusions

Identifying and characterizing particles that act as ice nuclei in mixed-phase clouds is important for an understanding of the ice-forming processes. In order to identify effective INPs during field campaigns, one approach is to sample only crystals with a maximum diameter of 20 μm, in order to exclude riming, which is predicted to take place in crystals larger than 50 μm ice [22]. However, the no-scavenging assumption of crystals up to 20 μm is contestable since laboratory experiments and ice crystal sampled at Dome C have shown that even small ice crystals can scavenge aerosol even in the early stage of growth.

Another method adopted to identify the INPs in mixed clouds consists of comparing the chemical composition of the residual particles in small ice crystals with aerosol near the cloud. However, comparison of the aerosol particles in small ice crystals and in background aerosol conducted at the Jungfraujoch station gave contradictory results when evaluating the ice nucleation efficiency of soot/carbon aerosol particles in immersion freezing mode. As immersion freezing nucleation in mixed clouds follows several steps, including water vapour diffusion and scavenging processes, we suggest that environmental parameters influencing the nucleation processes should also be considered.

In conclusion, identifying effective INPs is problematic since even small crystal can contain not only the effective nucleating particles, but also scavenged particles.

After initial growth ice crystals can continue to grow by water vapour diffusion. Laboratory experiments confirm the contribution of diffusiophoresis with Stefan flow in the scavenging by snow crystals up to 3 mm in diameter. Once ice crystals exceed a certain size they grow by collision with supercooled drops that freeze (riming) or aggregate with other ice crystals to form snowflakes [1]. Rimed snow contains higher concentrations of aerosol than unrimed snow formed by

diffusional growth [55].

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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