A Review of Ice Particle Formation Models

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***Abstract*—Modelling of ice clouds in the atmosphere is in general a more complex task than modelling their liquid water counterparts, owing to the plethora of ice crystal nucleation processes and their non-linear dependence on surrounding conditions. Accurate modelling of ice clouds plays an important role in weather prediction and climatology, particularly in their contribution to greenhouse effect, global warming and precipitation processes, and the impact of aviation on the environment. In this paper, we review different ice particle formation models, focusing on the underlying assumptions, advantages and limitations of each scheme.**

***Index Terms*—cirrus clouds; climate modelling; heterogeneous nucleation; homogeneous nucleation; ice nucleation; ice particle; ice supersaturation; ISSR**

I. INTRODUCTION

A quick glance at the clouds above often decides whether to carry an umbrella to work. Clouds play a major role in weather prediction and climate monitoring. In particular, cirrus clouds that are made up of ice crystal particles contribute to the Earth's radiation budget and climate, and their effect on global warming is a topic of current research. Modelling ice particle formation and the continuously changing intricate microstructures within these clouds is indeed a challenging task.

A crucial process in our understanding of cloud particle formation is the initiation of a new phase, i.e., transitions of water from vapour to liquid or solid phases. The rates of 'vapour-to-liquid' and 'vapour-to-ice' phase transitions are determined by vapour supersaturations with respect to liquid or ice, respectively [1]. According to the glossary of Arctic climatology and meteorology by the National Snow and Ice Data Center, saturation is defined as the condition in which the partial pressure of any fluid constituent (water in the atmospheric air) is equal to its maximum possible partial pressure under the existing environmental conditions, such that any increase in the amount of that constituent will initiate within it a change to a more condensed state [2]. At saturation, the relative humidity (RH) is defined to be 100%. An RH value greater than this corresponds to supersaturation. Saturating water vapour pressure over ice is less than that over water, and therefore supersaturation with respect to ice is attained earlier than with respect to water. Beyond a critical supersaturation or supercooling level, new phase formation (nucleation) takes place spontaneously due to spatial and temporal fluctuations in pressure and density [3]. Existing particle formation models differ primarily in their assumptions regarding the nucleation mechanism and ambient conditions.

Ice crystals are formed in the atmosphere around particles that act as nuclei. This physical process of ice nucleation is described very simply by Vali [4]. According to him, nucleation involves a spontaneous phase transition through *germs* of the new phase. In *homogeneous* nucleation, germs are isolated metastable clusters of the new phase that are distributed in the parent phase, and continually accrete and discard molecules. Accretion results in decreasing the germ surface-to-volume ratio, until it attains a *critical size* for stability. *Heterogeneous* nucleation differs in that germs are attached to a pre-existing structure (such as a solid surface), and requires less supercooling or supersaturation. Examples of nucleation processes are *deposition* (ice from vapour) and *freezing* (ice from liquid). Hetero- and homogeneous freezing and heterogeneous deposition are the ice nucleation mechanisms that occur in the atmosphere. Their relative roles are depicted in Figure 1.

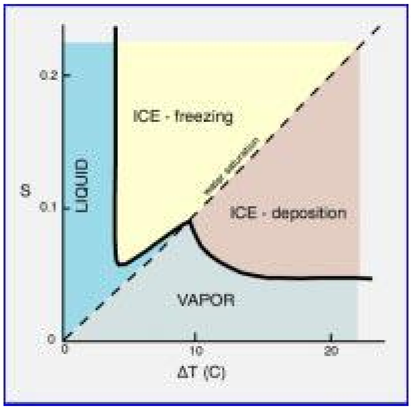

Figure 1. Deposition and freezing thresholds. S and $\triangle T$ denote supersaturation (with respect to ice) and supercooling, respectively. [4]

II. HOMOGENEOUS ICE NUCLEATION

The formation of cirrus clouds in environments with moderate to strong upward draughts or few germs is dominated by homogeneous nucleation, as elaborated by Tompkins *et al.* [5]. The relative humidity with respect to ice increases up to a critical threshold RH_{crit} of supersaturation as high as 150%, before nucleation begins under conditions of low temperature and large difference between the vapour pressures of liquid water and ice saturation.

At RH_{crit}, homogeneous nucleation takes place, and the accretion of water vapour molecules causes the RH to drop down to nearly 100%, as depicted in Figure 2(a). The model by the Integrated Forecast System (IFS) of the European Centre for Medium-Range Forecasting (ECMWF) shown in Figure 2(b) assumes supersaturation to be transformed into ice crystals immediately, by using a saturation-adjustment-type scheme [6].

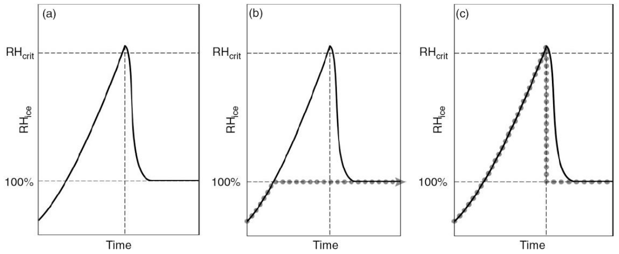

Figure 2. (a) RH evolution in a hypothetical parcel (grid cell) subject to adiabatic cooling at $T < 235$ K. (b) ECMWF IFS approximation. (c) Ice supersaturation parameterisation by Tompkins *et al.*. The dotted lines in (b) and (c) indicate approximations to (a). [5]

1) In pure ice phase, supersaturation is permitted in the clear-sky portion of the grid cell.
2) Ice nucleation can begin when the RH with respect to ice saturation locally reaches $RH_{homo} = 258.3 - \frac{T}{2.078}$.
3) However, since it is assumed that humidity fluctuations in the clear-sky portions are uniformly distributed with a fixed constant variance, ice nucleation initiates when the grid-mean RH exceeds a threshold that is lower than RH_{homo}.
4) In the presence of ice, deposition compared to the model time-step is fast enough to be approximated to exactly saturated conditions within the cloud, by diagnostic adjustments.

This scheme is, however, impaired by dry biases in the higher troposphere and inference of steadier values of humidity in time and space than reality. The model proposed by Tompkins *et al.* [5] addresses this limitation (Figure 2(c)) by including the effect of the nucleation process, in accordance with the following assumptions:

1) In cirrus cloud-scale models, a single freezing event cannot be extended over many time steps. The local ice saturation ratio is maximum at a critical value, beyond which freezing commences. It is assumed that nucleated particles appear immediately after this critical value is attained, i.e., when supersaturation locally reaches the critical value [7].
2) Sub-grid fluctuations of water vapour and temperature are neglected, i.e., humidity fluctuations are spread evenly over the clear sky, with uniform variance [8]. When the average RH of the grid cell in question rises above a limit that is lower than the local critical threshold for ice formation, nucleation sets in (not represented in Figure 2(c)).
3) In the presence of ice, a general-circulation model (GCM) time step is large compared to the deposition action. Instantaneous adjustments can be made to saturated conditions inside a cloud to resemble the deposition process, once nucleation is initiated.

An advantage of this model is that artificial horizontal transition of water vapour between clear and cloudy regions of the grid cell is not allowed. However, it is possible that no cloud may be present within a subsaturated grid cell, and the ice crystal concentration is not considered in these assumptions. This scheme has the effect of minimised high cloud cover (especially over the tropics) and increased upper troposphere humidity. Comparison with aircraft observations have indicated satisfactory predictions at RHs of up to about 130%, beyond which some underestimation occurs with this model. Validation with MLS humidity data retrievals resulted in satisfactory emulation of geographical position and upper tropospheric supersaturation frequency. A possible future improvement to this scheme is comparison to radiosonde network and application of an appropriate radiosonde bias correction [5].

III. HETEROGENEOUS ICE NUCLEATION

Modelling of nucleation on foreign surfaces faces the problem of surface characterisation, particularly of surface features that play the role of catalysts. This section deviates from cloud-scale model analysis, and deals with the small-scale physical processes that take place during heterogeneous nucleation. This serves as a basis for implementation of cloud-scale heterogeneous nucleation models, examples of which are discussed in the next section.

Wettability and reflection in the contact angle of a germ that is assumed to have a spherical cap shape on an insoluble substrate, are the fundamental phenomena used to describe heterogeneous nucleation, according to Vali [4]. This model is used for the formation of a liquid phase from vapour, as well as for solid from liquid. The critical size of the germ is determined by the germ radius. Using the germ geometry, free energy of germ formation and interfacial energy with respect to the substrate, the nucleation rate of ice on a substrate may be deduced. Considering the substrate to be a spherical solid, nucleation on aerosol particles can be modelled. Different geometrical germ shapes are assumed such as flat cylinders for crystalline germs for more accurate results, as shown in Figure 3.

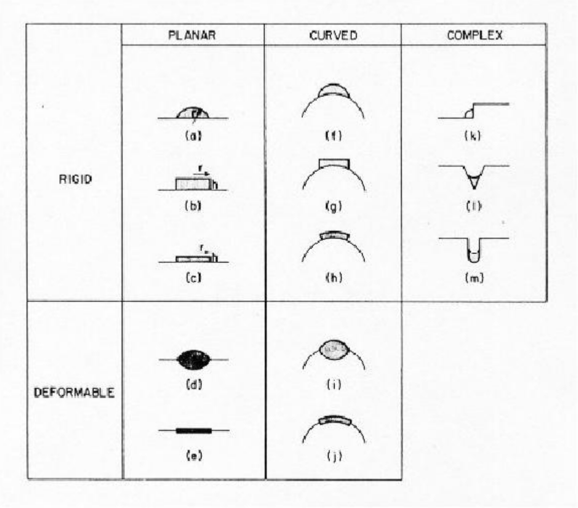

Figure 3. Some simple shapes of ice germs on solid or deformable substrates. [4]

Specific surface features have a higher probability of ice nucleation, especially deposition. However, the conditions at these features that enable them to initiate nucleation are not clearly known. Figure 4 shows photographs of ice crystals nucleated on various substrates. Some patterns set by the substrate are visible. Competition among growing crystals, and to a certain extent, chance determines where along a crystal substrate step nucleation is to take place. Studies have shown (Figure 5) that ice crystals often form in the same location sites, when a substrate is repeatedly exposed to supersaturation.

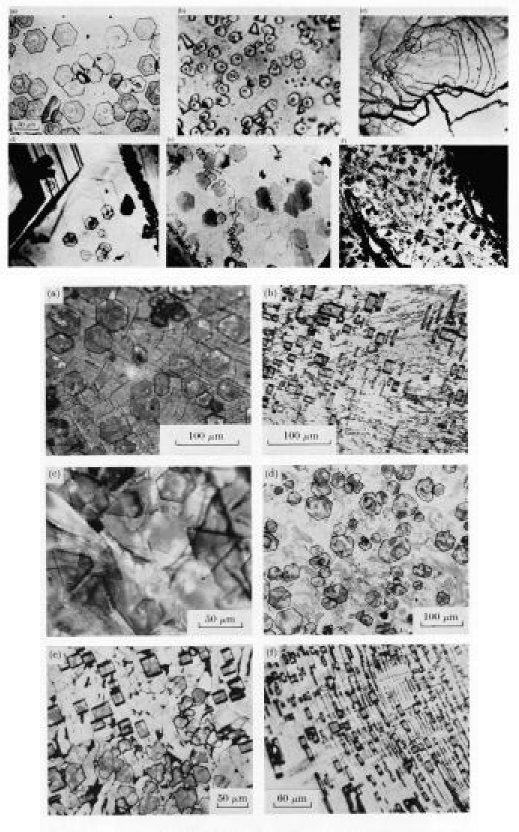

Figure 4. Ice crystals formed by nucleation from vapour on inorganic (top) and organic (bottom) substrates. [4]

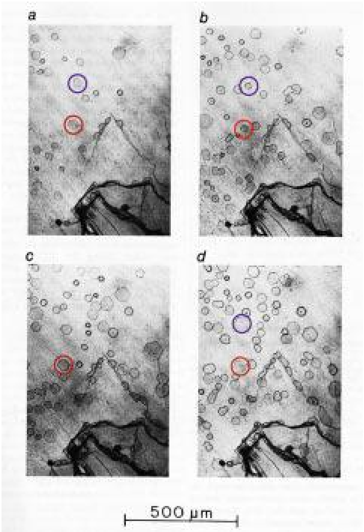

Figure 5. Repeated exposure of silver iodide at -14ºC to increasing supersaturations with respect to ice. Crystals nucleate in increasing numbers, but several sites activate repeatedly, such as the circled sites. Ice was completely evaporated between the exposures. [4]

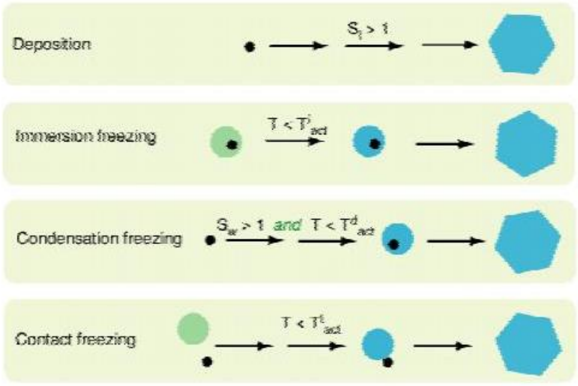

Figure 6. Ice nucleation modes in the atmosphere. T^i_{act}, $T^d_{act}$and T^t_{act} refer to activation temperatures for immersion, condensation and contact freezing respectively, and S denotes nucleation rate. [4]

According to the Vali model, there are at least four possible *nucleation modes* in the atmosphere, which are different routes taken by water vapour and nucleation-inducing aerosol to reach ice initiation. These are described schematically in Figure 6.

The nucleation abilities of different substrates in this model are expressed as a supersaturation or temperature needed for a specified rate of nucleation per unit surface area. Experiments with silver iodide have demonstrated its near-insolubility in water and likeness to the crystal structure of hexagonal ice. This lattice structure similarity and/or other factors are employed in qualitatively determining ice nucleation ability. Although models of lattice fitting in terms of interfacial energy have been proposed, quantitative small-scale estimations of heterogeneous ice nucleation remain inaccurate due to difficulties in bond energy determination. However, large-scale parameters such as ice crystal number densities and size distributions have been modelled, and are utilised in cirrus cloud models that take heterogeneous nucleation into account, as elaborated in the following section.

IV. CIRRUS CLOUD MODELS

Cirrus clouds may be formed at temperatures lower than 238 K, by homo- and heterogeneous nucleation mechanisms. According to Lohmann *et al.* [12], large and thin hexagonal plate ice crystals discovered in low concentrations near the tropical tropopause indicate the presence of particles that play the role of ice nuclei at low supersaturations with respect to ice. Mixed organic/inorganic aerosols can form glasses at upper troposphere temperatures, which could greatly lower their homogeneous nucleation capability. Additionally, ice growth impedances could potentially cause high supersaturations within cirrus clouds. Ice supersaturated regions (ISSRs) have gained importance in recent years as potential formation regions of cirrus clouds and persistent contrails, particularly in the upper tropical troposphere.

Lohmann *et al.* also describe a two-moment microphysics model, implemented in the ECHAM5 GCM. According to this model, cirrus clouds are formed by homogeneous freezing of solution droplets and heterogeneous freezing on immersed dust nuclei. ECHAM5-HAM, an extended version of ECHAM5, enables prediction of the aerosol mixing state apart from aerosol mass and number densities. A depositional growth equation is solved, which allows supersaturations with respect to ice. Thus, upon crossing a threshold, homogeneous freezing forms cirrus clouds. After cirrus formation, the whole grid cell is assumed to be cloudy. Between 0 and –35 °C, ice crystals that rapidly grow large in mixed-phase clouds are assumed to possess a plate-like shape, in contrast with prior spherical shape assumptions. However, the assumption of spherical shape is retained for small-sized crystals. Figure 7 shows a distribution of ice crystal shapes in the troposphere.

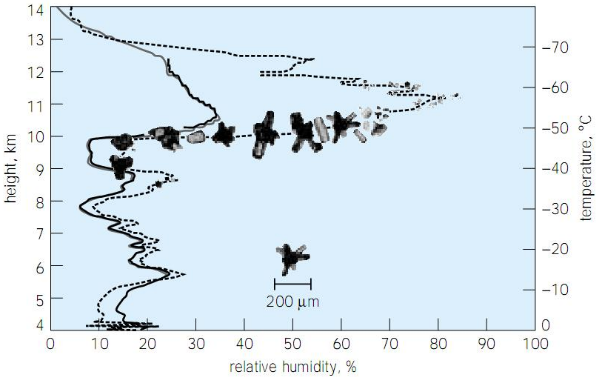

Figure 7. Size and shape of ice crystals, as a function of height, temperature, and relative humidity. Data captured by a replicator balloon sounding system in Marshall, Colorado, USA on November 10, 1994. The dashed and solid lines indicate the relative humidity measured by cryogenic hygrometers and Vaisala RS80 instruments, respectively. (Graphic by Andrew Heymsfield, National Center for Atmospheric Research) [13][14]

Although still not completely understood, it is known that cirrus cloud formation has intricate dependencies on many parameters, such as temperature, RH, wind fields, ambient aerosol, as well as large-, meso-, and small-scale dynamics. The scheme proposed by Spichtinger *et al.* [9] takes into account various scales of dynamics, and has been validated by extensive simulations.

1) The processes of homo- and heterogeneous nucleation, deposition growth/sublimation and sedimentation of ice crystals are implemented into two models (the box and EULAG models, explained below) with identical ice microphysics parameterisations, that can be used for different applications.
2) Prognostic equations are formulated for the number and mass distributions of various aerosol and ice classes, from which their sizes and shapes are determined. This bulk microphysical two-moment scheme implies that ice class differentiation is not done as cloud ice and snow, but based on nucleation processes or the respective aerosol classes. Ice classes are each linked to an aerosol type that freezes into the corresponding ice class.
3) Assumptions are made regarding the shape (hexagonal column) and terminal velocity (as a function of crystal mass) of single ice crystals, and *a priori* distribution type for masses of ice crystals and single aerosol particles (lognormal).
4) Homogeneous freezing of supercooled aqueous solution droplets and heterogeneous freezing on solid aerosol particles are parameterised using this scheme.

A simple zero-dimensional box model of the ice microphysics is used for substantiation and fast calculation of the nucleation parameterisation. This model consists of a packet of air whose motion in the vertical direction is governed by a velocity function over the entire simulation time, under the assumption of purely adiabatic processes. The Eulerian/semi-Lagrangian (EULAG) model is anelastic and non-hydrostatic, and is used for the tested microphysics. Its application covers different scales and dynamics, including stratified movement of air over elevations in landmass, and gravity waves caused by convection. Less diffusive advection schemes are a merit of this model. Dynamics that take into account water vapour, as well as thermodynamics are considered for incorporating cloud physics into this model. The box model does not require large computational capability, and hence the time step can be very small. This is not the case with the EULAG model, and an adaptive microphysical time step is used here.

Validation of this model is carried out in two steps.

1) The first step comprises of comparing the ice crystal number densities obtained from simulation runs of this box model under the same conditions as the corresponding box models by Kärcher *et al.* [7][11], along with some sensitivity studies. These box models contain a highly-resolved aerosol size distribution, used for testing an analytic relationship between the maximum possible number density of ice crystals formed by homogeneous nucleation and vertical velocity.
    a) Idealised simulations utilise only one class of ice formed by homogeneous nucleation. Nucleation is assumed to have no effect on the background aerosol number density, by assuming this density to have a very large value (~10000 cm^{-1}). This leads to two errors: underestimation of maximum nucleation rates, and clipped duration of the nucleation event. These can be mitigated by introducing prognostic equations for aerosol dynamics, and by reducing the average aerosol size during the nucleation phase. However, it was found that more realistic assumptions reduce the above errors, obviating the need for complicated correction measures.
    b) Realistic background conditions utilise aerosol concentrations that are much less than in the idealised simulations (~300 cm^{-1}). Nucleation thus affects the ice crystal number densities, and a correction is used for the mean aerosol mass, since the aerosol size distribution shifts to smaller masses after a nucleation event. As a result, the number of ice crystals formed may be limited by the available aerosol.
    c) Too long time steps yield large deviations from

reference cases at high vertical velocities. This may be attributed to the non-linear behaviour of nucleation and depositional growth. These deviations are larger at higher temperatures.

2) The deficiency of spatial dynamic phenomena such as sedimentation in the box model have led to extensive simulations using the EULAG model. This has demonstrated the influence of dynamics on cirrus cloud formation and development [10]. The second step is therefore a performance comparison between 1D simulations of the EULAG model and spectrally resolving schemes for Arctic cirrostratus triggered by a constant upward draught. Interesting effects were observed as a result of the interplay between crystal growth and sedimentation. In the lower parts of clouds, the RH_{crit} required for homogeneous nucleation is not reached, since crystal growth reduces supersaturation. However, falling ice crystals are formed in the upper parts of clouds which attract water vapour. Ice crystal number densities taken over the entire cloud are relatively low, despite continuous crystal formation by homogeneous nucleation. Hence, supersaturation is preserved within the cloud.

Sedimentation enacts a vital role in the development, structure and preservation of supersaturation in clouds. In case of little or no sedimentation, nucleation would result from cooling of the supersaturated zone, and crystal growth would occur until the extra vapour is absorbed over the supersaturated zone. These results are depicted in Figures 8 and 9. The results of simulation runs with and without sedimentation are reproduced in Figure 10.

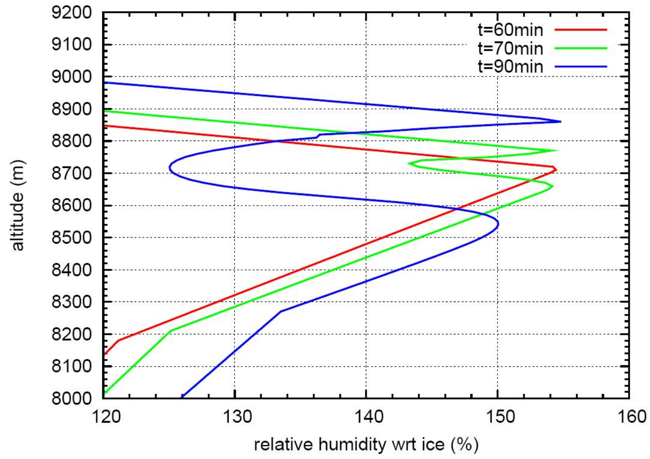

Figure 8. Temporal evolution of vertical RH profiles for 60-90 min simulation time, with a constant uplift of 0.06 ms^{-1}. Ice crystal growth causes the notch. Cloud top is supersaturated. [9]

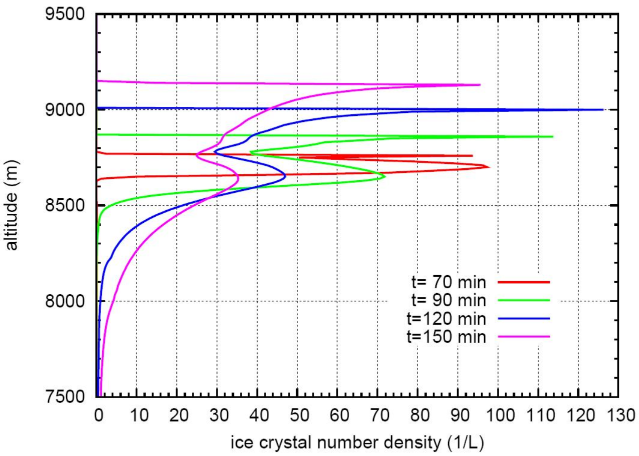

Figure 9. Downward-moving peak of high ice crystal concentration formed by first homogeneous nucleation event at about 60 min, with a constant uplift of 0.06 ms^{-1}. High peak values of number densities at cloud top indicate continuous nucleation. [9]

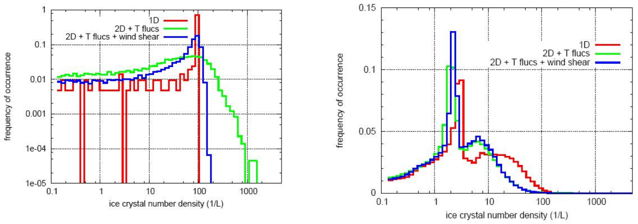

Figure 10. Three reference simulations (red: pure 1D, green: temperature fluctuations, blue: temperature fluctuations plus wind shear) (a) without sedimentation, and (b) with sedimentation, with an uplift of 0.05 ms^{-1}. [10]

The ice crystal number density disperses over the entire cloud due to sedimentation, and therefore, the frequency of occurrence shifts towards lower densities. However, it may be noted that sedimentation has a lesser impact if the time scale of sedimentation is larger than that of ice crystal growth. Box modelling of clouds is usually applied during the formation and early development stages of clouds, when sedimentation has a weak effect. Additionally, small-scale fluctuations may have drastic effects on cloud evolution and structure, especially owing to the non-linear processes such as nucleation that are involved. Future extensions to this model include a 2D approach using temperature fluctuations, and investigating the effects of different nucleation mechanisms and orographic gravity waves on cirrus cloud formation and development.

V. CONCLUSIONS

Ice formation in cirrus has been analysed in great detail by a number of authors, in terms of homo- and heterogeneous nucleation processes, deposition and freezing. Several models have been proposed to simulate cirrus cloud formation under various conditions of temperature, pressure, environment and altitude. These models differ mainly in their assumptions on the nucleation mechanism and meteorological conditions. The processes of deposition and freezing form the essence of ice particle formation. Other modes merely serve to alter the conditions of the nucleating particle, or change the interaction between the particle and the supercooled liquid droplet. Several other aspects of ice nucleation deserve discussion, but they would lead to details beyond the aim of this paper.

Despite considerable understanding in the geophysical research community on ice particle formation, there is need for establishing further atmospheric relevance to these concepts, by extensive observation and experimentation. These methods have demonstrated the crucial role cirrus clouds play on the Earth's climate, particularly the greenhouse effect. However, data collected from observational satellites are not yet sufficient to construct an exhaustive climate model that determines the impact of long-term variability of cloud cover, height and composition on the Earth's temperature and precipitation patterns. There is also need to improve radiometer technology and retrieval techniques, to enable detection of thin and subvisual cirrus clouds. Modelling ice particle and cirrus cloud formation is thus an intriguing problem, and requires considerable interplay between observational and theoretical research towards the end of an accurate Earth climate model.

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