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Cirrus Clouds and Climate Engineering: New Findings on Ice Nucleation and Theoretical Basis

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1. Introduction

Geo-engineering, henceforth referred to as climate engineering, can be described as an intentional intervention on the Earth's climate system for the purpose of temporarily reducing the increase in surface temperatures due to global warming. While it is generally acknowledged that climate engineering cannot solve the problem of global warming resulting from unrestricted greenhouse gas (GHG) emissions (Lauder and Thompson, 2010), it may "buy time" for non-carbon energy systems to dominate global energy production and for GHG emissions to be reduced to safe levels. The longer meaningful global policy decisions on GHG emissions are delayed, the stronger the demand is likely to be for climate engineering research and development. As described by Anderson and Bows (2010), even if an effective global treaty on GHG emissions were to take effect now, it would still be extremely difficult to prevent mean global surface temperatures from rising above 2°C (above which it is argued that the consequences of climate change would be unacceptable). It is thus defensible to argue that some combination of GHG mitigation and climate engineering may be necessary to limit the mean increase in global surface temperatures to 2°C.

A modeling study by Arora et al. (2011) also finds that a mean global warming exceeding 2°C may be unavoidable. Using updated GHG emission scenarios and an upgraded Earth system model (which accounts for aerosol effects and represents the carbon-cycle more realistically), the study finds that even under the lowest, most optimistic GHG emission scenario, the global average temperature will still increase more than 2°C (the limit agreed to by various governments in the Copenhagen accord) by 2100. To limit warming to 2°C by 2100, carbon dioxide emissions would need to be reduced to zero over the next 50 years followed by ongoing carbon sequestration (removal of CO₂ from the atmosphere) through the end of this century. While few scientists advocate tinkering with the earth's climate (Schneider 2010), climate engineering of some kind may become a necessity rather than an option.

Two types of climate engineering have been recognized: carbon dioxide removal (CDR) strategies and solar radiation management (SRM). CDR approaches can be biological, such as iron fertilization of the oceans to increase phytoplankton uptake of CO₂ (Lampitt et al., 2010; Smetacek and Naqvi, 2010), or they can be physically based, such as the direct capture

of CO₂ from the atmosphere using chemically treated scrubbers with subsequent carbon sequestration or conversion into carbon-neutral fuels (e.g. Keith et al., 2010). CDR is arguably the best climate engineering approach as it could potentially stabilize atmospheric CO₂ concentrations at safe levels and prevent ocean acidification. Unfortunately, iron fertilization is unlikely to remove CO₂ from the atmosphere at rates sufficient to prevent future warming, and air capture would likely require decades before such devices are numerous enough throughout the world to make a significant impact.

SRM refers to efforts that increase the reflection of solar radiation to space, thus reducing surface temperatures. A major drawback common to all SRM methods is that they do not address the problem of ocean acidification. The most studied SRM method is to reflect incoming sunlight with sulfuric acid aerosol injected into the stratosphere (similar to how volcanic aerosols cool climate). Although this method may have serious drawbacks such as a likely interference with the hydrological cycle (producing drought in some regions), ozone destruction and turning the sky white, this SRM method appears capable of rapidly neutralizing the warming due to a CO₂ doubling (Lenton and Vaughan, 2009), and appears relatively easy and quick to deploy. Another SRM method that might neutralize the warming from a CO₂ doubling is the brightening of marine stratus clouds (Latham 1990, 2002; Latham et al., 2008, Lenton and Vaughan 2009). These clouds are made more reflective by enriching the marine boundary layer with abundant cloud condensation nuclei (CCN) produced by a fine spray of sea water from specially designed boats (Salter, 2008). Potential drawbacks include regional changes in sea surface temperatures producing changes in ocean circulation patterns and modification of regional weather systems (The Royal Society, 2009).

A conceptual shortcoming of SRM-based methods is that they implicitly address a problem of reduced heat output (GHGs trapping thermal radiation) by reducing radiation input (reflecting sunlight to space). A third climate engineering approach recently considered (Mitchell and Finnegan, 2009; henceforth MF2009) is to directly address the problem of increased troposphere heat content by increasing its heat output (i.e. shifting its thermal equilibrium in favor of cooler temperatures). If it were possible to modify the troposphere's heat balance to reduce surface temperatures, this would offset the effect of heat-trapping GHGs in a more direct fashion. This third approach might be called earth radiation management (ERM). Under this definition, reducing GHG emissions and CDR are in effect ERM methods.

The purpose of this chapter is to describe an ERM approach pertaining to cirrus clouds and to test this hypothesis against new, more reliable cirrus cloud measurements. Like the SRM methods, this ERM method does not address ocean acidification, and effective GHG mitigation strategies along with possibly CDR methods would be needed to responsibly address the problem of global warming. This chapter is organized as follows. Section 2 provides an overview of the cirrus cloud climate engineering idea, which is described in greater detail in MF2009. Section 3 presents new research findings that test the conceptual foundation of this ERM approach. These findings are not only relevant to climate engineering, but are relevant to basic research on cloud-aerosol-climate interactions as well. Section 4 addresses the potential of over-cooling the planet and the need for climate monitoring. Some possible social, political and economic ramifications of climate engineering are discussed in Section 5, and a summary with conclusions is given in Section 6.

2. Modification of cirrus clouds to reduce global warming

In a study involving 1000's of "perturbed physics" global climate model (GCM) simulations using the U.K. Hadley Centre GCM, Sanderson et al. (2008) identified physical processes responsible for changing climate sensitivity (i.e. the equilibrium response of global-mean surface temperature to CO₂ doubling). The most important process affecting climate sensitivity was entrainment associated with deep convection, which strongly affects water vapor amounts in the upper troposphere. The second most important process was the ice fall speed from cirrus clouds, which affects the cirrus cloud life cycle and cloud coverage, the cirrus cloud optical depth and the upper troposphere water vapor. These findings imply that upper troposphere water vapor and cirrus clouds are having the greatest impact on climate sensitivity. Thus, an effective ERM strategy might target these components of the climate system which regulate outgoing longwave radiation, or OLR.

A modeling study by Mitchell et al. (2008), using an atmospheric GCM known as the Community Atmosphere Model version 3 or CAM3, also found that the CAM3 climatology was sensitive to the ice fall speed which had a profound influence on cirrus cloud cover, ice water path, optical depth and related radiative fluxes and heating rates. In addition, this study demonstrated a substantial dependence of the ice fall speed on the concentrations of relatively small ice crystals in the ice particle size distribution (PSD), thus suggesting a dependence of the representative fall speed on ice crystal nucleation rates. This suggestion will be explored further in this chapter.

Cirrus clouds are similar to GHGs in that they trap OLR by absorbing/emitting upwelling thermal radiation back towards the surface, which has a warming effect on climate. This "greenhouse" effect of cirrus clouds becomes stronger the higher they get since the effective temperature at which the Earth radiates thermal energy to space depends on the temperature of the overlying clouds. An abundance of cirrus will cause this effective temperature to be relatively low (similar to the temperature of the cold cirrus clouds). This reduces the OLR and traps thermal radiation below the cirrus clouds that would otherwise escape to space. In addition, cirrus clouds also reflect sunlight back to space (a cooling effect on climate), but this albedo effect is less efficient than their "greenhouse" effect. Thus, cirrus clouds have a net warming effect on the earth's climate (Hartmann et al., 1992; Chen et al., 2000). It follows then that the most effective way to increase OLR may be to reduce the cloud cover of the highest, coldest cirrus clouds.

One method for how this goal might be accomplished is described in MF2009. They suggested "seeding" cirrus clouds with efficient ice nuclei that would out-compete the natural ice nuclei for water vapor. Aerosol particles that serve as ice nuclei often "activate" to form ice crystals at some threshold supersaturation with respect to ice. Natural aerosols that serve as ice nuclei activate at relatively high thresholds compared to those required for efficient ice nuclei such as silver iodide (used in cloud seeding operations). Introducing efficient ice nuclei into cirrus clouds at relatively low concentrations should then preclude the activation of most natural ice nuclei since the efficient ice nuclei (i.e. the cloud seeding aerosol) will activate and grow ice crystals at lower supersaturations and their growth will prevent water vapor concentrations from ever reaching the threshold supersaturation needed to activate the natural ice nuclei. This has a two-fold effect of depressing water vapor concentrations in supersaturated environments and producing fewer ice crystals. While lower in concentration than the natural ice nuclei, the concentration of the cloud seeding aerosol should be sufficient to prevent the ice supersaturation from rising

significantly as an air parcel ascends. This will insure that primarily only the seeded ice nuclei will produce ice crystals and that these crystals will grow to larger sizes than ice crystals produced by the more numerous natural ice nuclei. That is, a given amount of cloud condensate distributed among fewer ice crystals results in larger ice crystals. These larger, more massive ice crystals will have larger fall velocities than the natural ice crystals. As noted above, higher fall speeds will result in shorter cloud lifetimes, less cloud coverage, lower ice water paths and lower cloud optical depths. This would allow more OLR to escape to space.

2.1 Ice nucleation in cirrus clouds

Ice nucleation mechanisms can be grouped into two categories: (1) heterogeneous nucleation processes and (2) homogeneous freezing nucleation. The first category includes the mechanisms of deposition nucleation, contact nucleation, immersion freezing nucleation, condensation freezing nucleation and others (for a description of these mechanisms, see Pruppacher and Klett, 2010). These processes are referred to as heterogeneous since they involve a substrate other than water (vapor or liquid) to initiate freezing. The substrate could be mineral dust, black carbon aerosol or virtually any substance that promotes the freezing of supercooled liquid water. The rate of ice crystal production from these heterogeneous mechanisms is markedly slower than the production rate from homogeneous freezing nucleation (Pruppacher and Klett, 2010). Thus ice crystal concentrations produced through heterogeneous nucleation processes are lower than those produced through homogeneous freezing nucleation. In contrast to heterogeneous nucleation, homogeneous freezing nucleation does not require a substrate to initiate freezing. It occurs when a soluble aerosol particle (e.g. ammonium sulfate or nitrate, sulfuric acid, or any soluble hydrophilic substance) accretes water vapor in a humid environment to form a haze droplet (below water saturation), whereby the particle is dissolved resulting in a homogeneous solution. When the temperature is less than $\sim -38^{\circ}\text{C}$ and the relative humidity with respect to ice (RH_i) is greater than $\sim 140\%$, these haze droplets spontaneously freeze forming ice crystals. For the details of this homogeneous nucleation process, see, for example, Sassen and Dodd (1988), Heymsfield and Sabin (1989) and Koop et al. (2000). Kärcher et al. (2007) provide a discussion of ice nucleation processes relevant to cirrus clouds.

2.2 Reducing cloud cover for the coldest cirrus clouds

Cirrus clouds might be modified by seeding aerosol at any temperature, provided the natural ice nuclei, whether heterogeneous or homogeneous, activates at a RH_i well above the RH_i threshold of the seeding aerosol. For example, if the RH_i threshold for natural ice nuclei is $\sim 120\%$ and the threshold for seeding aerosol is $105\% \text{RH}_i$, the cirrus modification process described above should be possible. For homogeneous freezing nucleation, the RH_i threshold is greater than 140% (e.g. Kärcher et al 2007), making it even more likely that the seeding aerosol will out-compete the natural ice nuclei for water vapor. Moreover, homogeneous freezing nucleation requires temperatures colder than -37.5°C (Rosenfeld and Woodley 2000), making this temperature regime ideal for cirrus modification provided that homogeneous freezing is an important nucleation mechanism in that temperature regime. On this question there is considerable controversy, and one of the aims of this chapter is to provide new evidence towards resolving this controversy. If homogeneous nucleation is an important nucleation process for temperatures less than -40°C , then cirrus clouds at such

temperatures should be most susceptible to modification as described above. Forming in an air mass conditioned with seeding aerosol at the right concentration, cirrus ice crystals should grow larger and fall faster, depleting the air mass of moisture and reducing cirrus cloud coverage over time. As noted above, the most effective way to increase OLR may be to reduce the cloud cover of the highest, coldest cirrus clouds, and it appears that these cirrus clouds could be the most susceptible to modification.

2.3 Reducing upper tropospheric water vapor

To increase ice crystal fall-speeds, the direct aircraft seeding of cirrus may have the opposite effect (to decrease fall-speeds), since the seeding aerosol is initially concentrated upon release from an aircraft and may produce many relatively small ice crystals. Rather, seeding aerosol should be released in the upper troposphere under clear conditions and allowed to become well dispersed, conditioning a vast air mass with seeding aerosol that has a concentration range that promotes large ice crystal growth. Then under the right conditions cirrus clouds will form in this clear air having relatively large ice crystals and thus a relatively short life-cycle.

Vast regions in the upper troposphere are supersaturated with respect to ice yet remain cloud-free (Helten et al. 1998; Read et al. 2001; Spichtinger et al. 2003, 2004). The above seeding strategy would thus initially produce more cirrus clouds since the seeding aerosol would have a much lower RH_i threshold than the natural ice nuclei, forming cirrus clouds in these supersaturated regions. However, this effect should be transient since over time, the relatively large ice crystals would sediment to lower levels and warmer temperatures where the cirrus greenhouse effect is less. Water vapor concentrations in the upper troposphere should decrease as this moisture is exported to lower levels, decreasing the water vapor greenhouse effect. Indeed, this is what was observed in two GCM experiments when the ice fall-speed was increased (Lohmann et al. 2008, MF2009). The globally averaged clear-sky OLR was decreased by $\sim 0.7 \text{ W m}^{-2}$ in the Lohmann et al. study (MF2009) due to small changes in the ice fall-speed.

2.4 GCM studies

In MF2009, two GCM studies (Lohmann et al. 2008 and Mitchell et al. 2008) are discussed that lend credibility to this cirrus engineering idea. The study by Lohmann et al. contrasted ECHAM5 GCM simulations based on homogeneous freezing nucleation with simulations based on either heterogeneous nucleation or a combination of homo- and heterogeneous nucleation. This study indicated that a heterogeneous nucleation threshold of $RH_i = 130\%$ would increase effective ice crystal size by $\sim 11\%$, and that this would increase the ice fall-speed enough to produce a net global cooling of $\sim -2.7 \text{ W m}^{-2}$, with 2 W m^{-2} coming from cirrus cloud reduction and 0.7 W m^{-2} due to water vapor reduction. This compares with the radiative forcing due to a CO_2 doubling of 3.7 W m^{-2} . No GCM experiments have been conducted using very efficient ice nuclei ($RH_i \sim 105\%$), and thus greater increases in ice crystal size and fall speed, and greater cooling effects, might well be possible.

The study by Mitchell et al. (2008) did not examine the impact of different ice nucleation mechanisms on cirrus cloud radiative forcing, but it did look at the impact of varying the concentration of small ice crystals in the PSD, which affects the mass-weighted ice fall-speed. Ice fall-speed differences between simulations were most significant at temperatures less than -45°C . The higher fall-speed simulation (lower concentrations of small ice crystals)

had 5.5% less cirrus cloud coverage on a global average. In the mid-latitudes and polar regions, shortwave cloud forcing was changed very little while longwave cloud forcing decreased up to 6 W m^{-2} (i.e. OLR increased by up to 6 W m^{-2}) in the higher fall-speed simulation.

2.5 Possible seeding strategies

This seeding could be done continuously on a global scale using commercial airliners to sustain a quasi-constant concentration of seeding aerosol in regions frequented by airliners. Commercial airliners typically fly at temperatures between -45 and -60°C , which is the temperature region of interest. If the seeding material was soluble in jet-fuel, the jet-fuel could be doped with seeding material, but extensive tests would need to be conducted to insure that burning the doped fuel had no detrimental effect on the jet engines. Alternatively, a flammable solution of seeding material could be injected into the hot engine exhaust.

Another approach would be to develop a fleet of unmanned drone aircraft to disperse the seeding material. While perhaps more expensive, this approach would not face the many legal and technical issues dealing with public safety on commercial aircraft. The lightweight drone aircraft would be relatively fuel-efficient and could be directed wherever seeding was needed, remaining in flight for extensive periods.

Bismuth tri-iodide (BiI_3) has been suggested as a possible seeding material (MF2009) since it is as effective as silver iodide (AgI, the most effective seeding agent known) at temperatures colder than -20°C , is $1/12^{\text{th}}$ the cost of AgI, and is non-toxic. Note that the active ingredient in Pepto Bismol, commonly used for treating diarrhea, is bismuth subsalicylate (525 mg per dose).

2.6 Advantages and drawbacks

Since the cirrus cloud engineering idea is new and untested in climate models, it is unknown what undesirable environmental consequences may exist regarding its implementation. As stated above, it does not address the problem of ocean acidification. However, a number of advantages are apparent: (1) it appears potentially capable of neutralizing the warming due to a doubling of CO_2 ; (2) if negative environmental consequences resulted from implementing the idea, the climate system should return to its pre-engineered state within months after termination of seeding (due to the fact that the seeding aerosol have an atmospheric residence time of 1-2 weeks); (3) unlike marine stratus clouds, cirrus clouds are generally present everywhere over the earth, making preferential cooling of certain regions less likely; (4) if GCM simulations indicate seeding only certain regions (such as polar and/or mid-latitude regions) renders the most favorable global climate conditions, then such seeding strategies could be adopted; (5) The use of non-toxic seeding aerosol does not appear to present an environmental hazard since conventional cloud seeding does not, with AgI concentrations in snow being less than 10 parts per trillion (Super 1986; Warburton et al. 1995). Moreover, this climate engineering approach does not appear to have the problems associated with stratospheric injection of sulfuric acid aerosol: (1) likely interference in the hydrological cycle, enhancing the frequency of drought in some regions; (2) ozone destruction and (3) turning the sky white. Clearly more research is needed to better understand the pros and cons of this approach.

2.7 Needed research

To determine whether this approach is viable, targeted research will be needed in the following areas:

1. Improved understanding of ice nucleation mechanisms in cirrus clouds and potential seeding aerosol
2. Small-scale field campaigns with research aircraft to provide limited testing of the climate engineering approach
3. Plume dispersion studies to determine the feasibility of establishing a background concentration of seeding aerosol suitable for favorable modification of cirrus clouds
4. GCM studies to determine the potential effectiveness and limitations of the climate engineering method, especially in regards to unwanted environmental consequences. The credibility of the results will depend on how realistically the cirrus clouds are represented in the GCM, including consistency between predicted and observed cirrus cloud coverage.

There are other areas of research and engineering that need to be explored for assessing the viability of this approach, but progress regarding the above research topics would be a good start.

A major uncertainty regarding this climate engineering approach is whether homogeneous freezing nucleation is an important process by which ice crystals are formed in cirrus clouds. Before GCM experiments can be taken very seriously, a basic understanding of the dominant ice nucleation mechanisms at temperatures less than -38°C is needed. While it appears possible to modify cirrus clouds when heterogeneous nucleation processes dominate (provided the difference in RH_i threshold is appreciable regarding natural and seeded ice nuclei), cirrus may be more easily modified if homogeneous freezing nucleation is active and an important process, since this indicates the RH_i threshold for natural ice nuclei is more than 140%, substantially above that of the seeding aerosol ($\text{RH}_i \sim 105\%$). In this case it should be relatively easy to modify cold cirrus clouds since the seeding aerosol should easily out-compete the natural ice nuclei for water vapor.

There is a large body of literature dealing with hetero- and homogeneous freezing nucleation in cirrus clouds with no clear verdict on what process dominates at temperatures less than -38°C . A recent study by Krammer et al. (2009) suggests that heterogeneous nucleation processes may dominate at temperatures less than -40°C based on RH_i measurements and ice particle concentration measurements in cirrus clouds. A common problem to this and many other studies addressing this nucleation issue from a measurement angle is that the ice PSD has only recently been measured with reasonable accuracy. Earlier measurements were plagued with the problem of ice particle shattering, whereby ice particles sampled by a probe impact and shatter on the inlet tube, producing anomalously high concentrations of small ice crystals (McFarquhar et al. 2007, Jensen et al. 2009, Lawson et al. 2010, Mitchell et al. 2010, Zhao et al. 2011). The subsequent sampling of these artifact ice fragments along with natural ice particles made it virtually impossible to say anything about ice nucleation processes in the atmosphere based on aircraft probe measurements. Recently PSD measurement probes have been improved to greatly reduce the shattering problem (Lawson et al. 2006a, Jensen et al. 2009, Lawson et al. 2010, Korolev et al. 2011, Lawson 2011). These relatively recent PSD measurements are discussed below in relation to several field campaigns sampling cirrus clouds. Moreover, these measurements provide greater insight into the role of homogeneous freezing nucleation in cirrus clouds and the viability of this climate engineering idea.

3. New findings

This section presents new in situ measurements regarding the role of homogeneous freezing and heterogeneous nucleation processes at temperatures less than -38°C . For tropical cirrus clouds, homogeneous freezing nucleation may dominate ice production under the higher updraft conditions associated with anvil cirrus clouds, based on a change in the PSD shape, ice particle number concentration/ice water content (IWC) ratio and the mean ice particle size near -40°C . For mid-latitude synoptic cirrus, homogeneous freezing nucleation appears to dominate ice production based on a marked change $\sim -40^{\circ}\text{C}$ regarding the PSD shape, the ice particle number concentration/IWC ratio, the mean ice particle size, the ice particle shape and the mass-weighted ice fall-speed. These results suggest that ice production in fresh anvil and mid-latitude synoptic cirrus clouds occurs at RH_i levels generally exceeding 140%, which should make these clouds easily susceptible to modification from seeding with efficient ice nuclei.

3.1 Field campaigns and measurement methods

The new findings reported here deal with the role of homogeneous freezing nucleation in cirrus clouds, based on field observations from primarily two field campaigns: (1) the Tropical Composition, Cloud and Climate Coupling (TC4) campaign, sampled in August 2007 near Costa Rica (funded by the USA's National Aeronautical and Space Administration or NASA), and (2) the Small PARTicles In Cirrus (SPARTICUS) campaign, with six months of sampling over the continental USA during the winter and spring of 2010 (funded by the U.S. Department of Energy, Atmosphere Radiation and Measurement (ARM) program). During TC4, the NASA DC-8 aircraft was used to sample fresh anvil cirrus (anvil cloud connected to column of deep convection), aged anvil cirrus (anvil cloud detached from column of deep convection), and in situ cirrus (not directly related to convection). The in situ cirrus appeared thin and tenuous in satellite imagery. This study uses TC4 flight data obtained on 22 July, 24 July, 5 August and 8 August 2007. On August 5th, the NASA WB57 joined with the DC-8 to vertically profile a deep anvil cirrus deck. To complement the in situ cirrus measured during TC4, in situ cirrus data from the NAMMA (NASA African Monsoon Multidisciplinary Analysis) campaign of 2006 were also used in this study for the following flight days: 19 and 26 August, 3 September 2006. Constant temperature transects or legs were flown through the clouds during TC4 and NAMMA, with one measured PSD representing one flight leg. No evidence of liquid water was found in the TC4 cirrus clouds. The SPARTICUS PSD used here are from 1-2 minute periods of flight time where the cirrus microphysical properties (median mass dimension and extinction coefficient) were not changing rapidly over time. A total of 174 SPARTICUS PSD were processed from 9 different days and 13 different flights, where only synoptic (non-anvil) cirrus was sampled. Using measurements from the Cloud Particle Imager (CPI), the relative humidity sensor, Forward Scattering Spectrometer Probe (FSSP) and the 2D-S probe, no evidence of liquid water was detected in these clouds.

In both field campaigns a relatively new probe was used for measuring the PSDs; the 2D-Stereo or 2D-S probe (Lawson et al., 2006a, Lawson 2011). The 2D-S directly measures ice particle length and projected area, and indirectly measures ice particle mass. The ice particle mass in each size bin was determined from 2D-S measurements of ice particle area using the mass-area relationship described in Baker and Lawson (2006). The IWC was calculated by integrating the PSD ice particle masses, and this method was shown to produce IWCs

consistent with those measured directly by the Counterflow Virtual Impactor (CVI) during TC4, with differences less than $\sim 50\%$ and an r^2 of 0.88 (Lawson et al. 2010; Mitchell et al., 2010). The CVI uncertainty in IWC has been estimated to be 13% at water contents of 0.05 to 1.0 g m⁻³ increasing to 16% at 0.010 g m⁻³ and to 40% at 0.0025 g m⁻³ (Heymsfield et al., 2007; Twohy et al., 1997, 2003). Thus the 2D-S yields direct or indirect measurements of the size distributions for ice particle concentration, area and mass. These three types of size distributions were used to calculate the mass-weighted fall speed, V_m , as described in Mitchell et al. (2011). As noted in Section 2, V_m is a critical parameter controlling the cloud life cycle and coverage, and associated radiative forcing.

As noted, historical PSD measurements in cirrus clouds have suffered from ice particles shattering on the probe inlet tube which artificially enhanced the concentration of small ($D < 100 \mu\text{m}$) ice crystals. The problem of ice particle shattering is greatly reduced in the 2D-S probe due to both probe design and the removal of shattered ice particles based on ice particle interarrival times (Lawson et al., 2006a; Jensen et al., 2009; Baker et al., 2009a,b; Lawson et al., 2010). Thus the PSDs measured during these field programs appear to be much more realistic than previous PSD measurements and render more realistic estimates of ice particle concentrations and V_m .

An additional capability of the 2D-S to make improved measurements of ice particle size and projected area is its ability to provide true 10- μm pixel resolution at jet aircraft speeds. In laboratory experiments the 2D-S probe was shown to accurately image an 8- μm fiber rotating at 233 m s⁻¹ (Lawson et al. 2006), which is greater than the true airspeeds that aircraft experienced during these field campaigns. Ice crystals smaller than this appear to have a negligible impact on most anvil cirrus optical properties (Jensen et al. 2009). This higher resolution improves the measurement of particle projected area.

3.2 Calculation of V_m

Perhaps for the first time, the mass weighted ice fall-speed or V_m was estimated directly from in situ measurements of ice particle area and mass, as described in Mitchell et al. (2011). This reduces uncertainty in V_m by eliminating intermediate steps or approximations used in its calculation. Since the ice particle mass is estimated from the 2D-S probe measurements (Baker and Lawson 2006), and the 2D-S IWCs compare favorably with the CVI IWCs, it appears that ice particle masses based on the Baker and Lawson area-mass relationship are realistic regarding the TC4 cirrus anvils. CVI measurements of IWC were made during the last week of the SPARTICUS campaign and have not yet been compared against 2D-S probe IWCs. Thus for now we assume that ice particle masses derived from the Baker and Lawson (2006) area-mass relationship are valid for SPARTICUS cirrus. The violation of this assumption could impose a systematic bias on our results regarding V_m .

With measurements of the concentration of ice particle area, estimated mass and number (to calculate mean bin values of area and mass) for each size bin of the 2D-S probe, these properties can be used directly to calculate the ice particle fall velocity in each size bin, using the fall speed formulation of Heymsfield and Westbrook (2010). The formulation of Heymsfield and Westbrook is more accurate for ice particles having aspect ratios far from unity, such as long columns and needles or “stellar” dendritic ice crystals.

The treatment of the mass-weighted PSD fall velocity, V_m , is calculated as:

$$V_m = \sum v(D)IWC(D) / IWC, \quad (1)$$

where $v(D)$ is the fall speed calculated for a given bin size D and $IWC(D)$ is the measurement derived ice mass concentration in a given 2D-S size bin. To calculate $v(D)$, the mean area $A_m(D)$ and mean mass $m(D)$ of the size-bins are needed, which are calculated from the measurements as:

$$A_m(D) = A(D) / N(D), \quad (2)$$

$$m(D) = IWC(D) / N(D), \quad (3)$$

where $N(D)$ is the bin number concentration and $A(D)$ is the measured projected area concentration in a given bin. In this way V_m is calculated as directly as possible from measurements. The V_m calculations reported in this study assume a reference temperature of -20°C and a pressure of 500 hPa.

3.3 Calculation of area ratios

The area ratio AR is the projected area of an ice particle divided by the area defined by a circle having a diameter equal to the particle's maximum dimension D . The AR is thus a metric for ice particle shape, with more branched or columnar ice crystals tending to have relatively low ARs whereas compact or quasi-spherical ice particles have relatively high ARs near unity. If there is a change in ice crystal nucleation mechanism $\sim -40^\circ\text{C}$, then this could be accompanied by a change in ice crystal shape. For this reason the AR was calculated for each size bin of the 2D-S probe. All measurements of $A_m(D)$ were checked to insure that $AR \leq 1.0$. In rare cases when $AR > 1.0$, then $A_m(D)$ was set to the area of a corresponding circle.

Since the AR is a function of ice particle shape, ice particle shape differences among PSDs were evaluated using an AR representing the entire PSD. This was done by summing $A(D)$ over the PSD for ice particles greater than $60\ \mu\text{m}$. While the 2D-S probe has $10\text{-}\mu\text{m}$ pixel resolution, ARs are more accurate when estimated for size $D > 60\ \mu\text{m}$. Moreover, smaller ice crystals tend to be quasi-spherical (Korolev and Isaac 2003; Mitchell et al. 2010; 2011). The same was done using ice spheres (i.e. ice particle maximum dimension defines a sphere from which the area cross-section is calculated). Then the area ratio of the PSD was determined as

$$AR_{\text{PSD}} = \frac{\sum_{D=60\mu\text{m}}^{D=D_{\text{max}}} A(D)}{\sum_{D=60\mu\text{m}}^{D=D_{\text{max}}} A_s(D)}, \quad (4)$$

where $A_s(D)$ is the area of a sphere having diameter equal to the ice particle maximum dimension and D_{max} is the maximum particle size of the PSD. Probability distribution functions (PDFs) of AR_{PSD} were produced for each type of cirrus cloud evaluated here to determine whether ice crystal shapes change near -40°C .

3.4 Tropical cirrus clouds: TC4 results

As noted, cirrus clouds sampled during the TC4 campaign were classified as either fresh anvils, aged anvils or as in situ cirrus. The PSDs from these clouds were grouped into temperature intervals of 5°C and mean PSDs for each temperature interval were calculated as shown in Fig. 1, which shows the temperature dependence of these mean PSD for each

cloud type. Since updraft speeds in deep convective clouds are relatively high (meters per second), ice crystals in fresh anvil cirrus were likely produced under relatively high updraft conditions. In aged anvil cirrus, no longer attached to their parent cumulonimbus cloud, updrafts should be much weaker or possibly not present. In situ cirrus clouds are formed through the gradual ascent of an air mass and thus ascending air motions in these clouds should be relatively weak. It is ascending air motions that cause air to become supersaturated and to produce a cloud, and the higher the updraft, the higher the supersaturation level. For this reason, homogeneous freezing nucleation may become more likely as the updraft increases since it requires $RH_i > 140\%$.

Figure 1 shows that for the fresh anvil cirrus there is a clear transition in PSD shape $\sim -40^\circ\text{C}$, with PSD colder than -40°C being approximately mono-modal in shape, and PSD warmer than -40°C being more bimodal in shape. This can be interpreted in terms of nucleation processes, with homogeneous freezing nucleation affecting the PSDs at temperatures less than -40°C . Homogeneous nucleation will produce ice crystals at a faster rate than

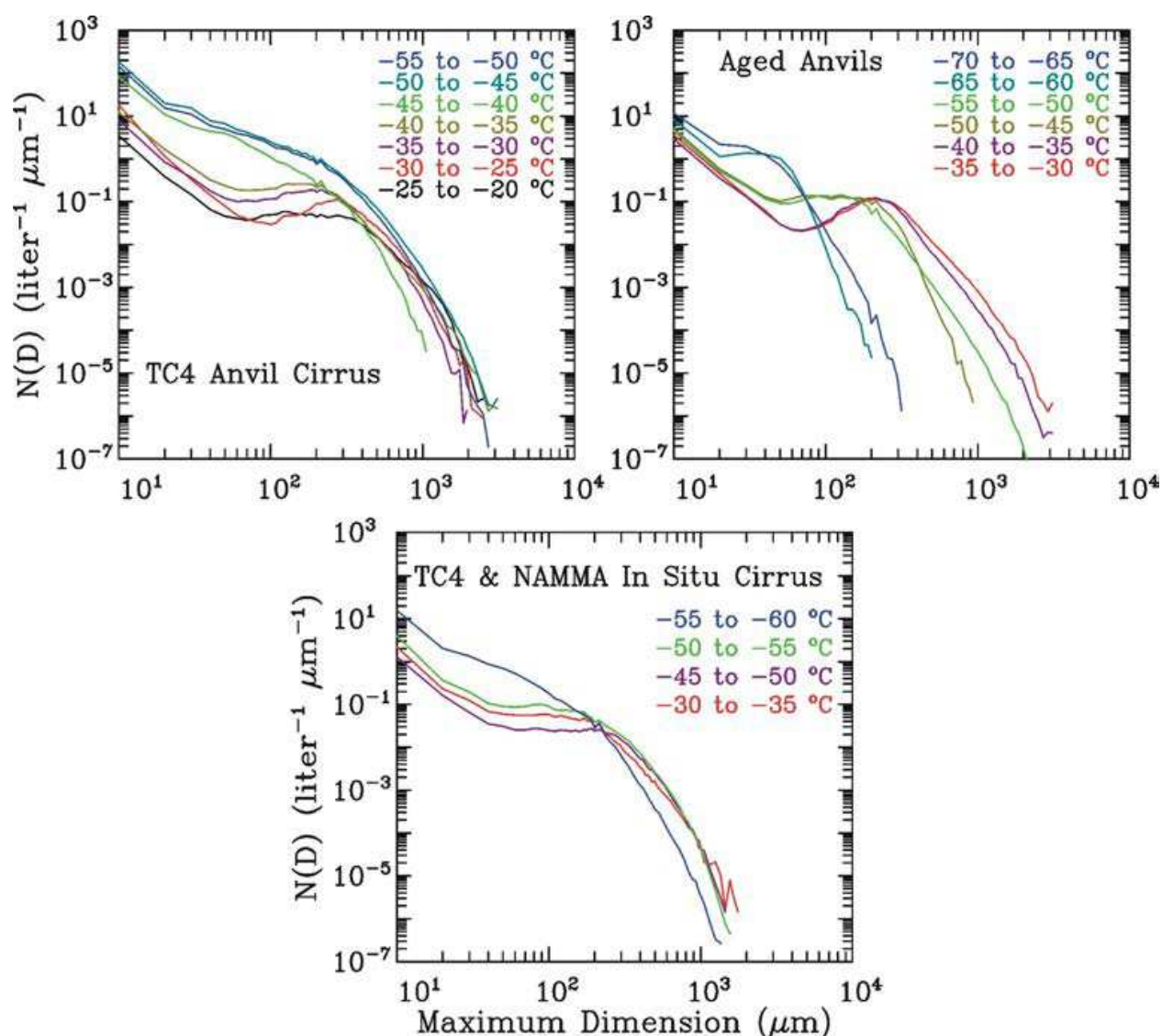


Fig. 1. Temperature dependence of mean PSD for three types of tropical cirrus clouds sampled during the TC4 and NAMMA field campaigns.

heterogeneous processes, thus elevating the concentrations of the smaller ice crystals relative to heterogeneous nucleation conditions ($T > -40^{\circ}\text{C}$). Ice particle growth processes also affect the PSD shape, such as aggregation. Aggregation is a process where ice particles collide with each other and combine to form larger “aggregates” comprised of individual ice crystals. It has been demonstrated that aggregation is an important growth process for ice particles in cirrus clouds (Mitchell et al. 1996). Aggregation will act on all PSD, depleting the concentrations of small ice crystals to form larger ones and possibly producing a secondary maximum in concentration $\sim 200 \mu\text{m}$ particle size when ice crystals are produced only through heterogeneous processes. This bimodal behavior apparently disappears when ice crystals are nucleated at a higher rate, building up the small crystal concentrations to produce approximately mono-modal PSD. Thus there is a counter-balancing effect between nucleation producing new small ice crystals and the removal of small crystals due to aggregation. When nucleation rates are relatively low (i.e. heterogeneous processes), aggregation effects should be more apparent, with bimodality developing. This explanation appears to fit the fresh anvil observations in Fig. 1 where updrafts are higher.

The temperature dependence of the fresh anvil cirrus could also be explained by a process known as size sorting, where larger ice particles fall faster than smaller ones, leaving higher concentrations of smaller ice crystals at higher cloud levels and building up the concentration of larger ice particles at lower levels. This process could explain the observed bimodality at warmer temperatures, or it could be a contributing factor, but this process alone cannot explain the transition observed at -40°C . Some may argue this transition at -40°C is a mere coincidence.

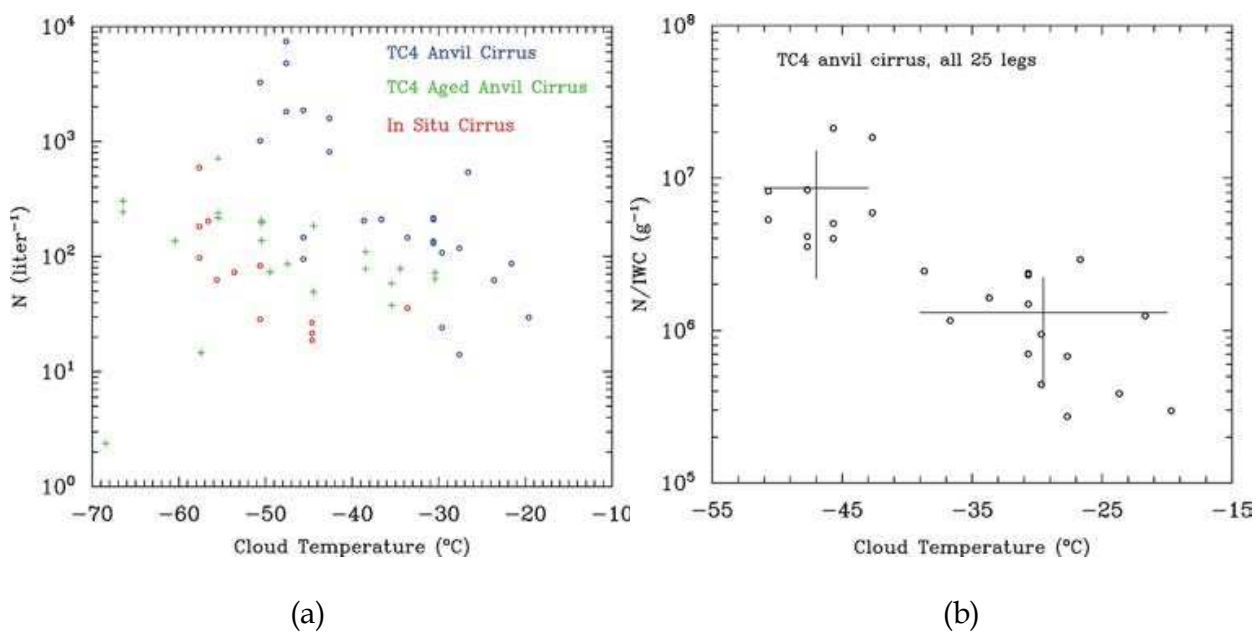


Fig. 2. a: Total number concentration for the three types of cirrus clouds sampled during the TC4 campaign. b: For the fresh anvil cirrus cases, shown is the total number concentration divided by the corresponding PSD IWC related to PSD temperature.

The aged anvil PSDs in Fig. 1 exhibit a similar temperature dependence, but without a distinct transition near -40°C . It is not clear whether this behavior is a “memory effect” from an earlier period when the anvil was “fresh” or whether homogeneous freezing nucleation was having an ongoing influence on the PSD temperature dependence. The PSD associated with in situ cirrus do not show a transition from bimodal to mono-modal size-spectra near -40°C , which appears consistent with their lower updrafts. The mono-modal PSD at the coldest level for in situ cirrus may reflect a reduced impact from aggregation near cloud top (aggregation influences on PSDs increase with distance from cloud top (Mitchell 1988, Mitchell et al. 1996)).

The above information can be presented in terms of the bulk properties of the PSD; total number concentration N and mean ice particle size (maximum dimension), \bar{D} . Figure 2a shows N for the three cirrus cloud types. The highest five N values are associated with an IWC greater than 0.3 g m^{-3} (very high for cirrus clouds), due to sampling closer to the center of convection. Since N partly depends on RH_i (related to the condensation rate and IWC), N was divided by the corresponding IWC to remove this dependence in Fig. 2b for the fresh anvil cirrus. This ratio is related to the nucleation rate coefficient J , which is the number of ice germs formed per unit volume (related to mass) of condensate per unit time. N/IWC can be thought of as a time integration of J in a cloud parcel, from ice nucleation onset to sampling time, although this is oversimplified as it ignores other processes such as aggregation and ice fallout. Since J for homogeneous freezing is known to be higher than J for heterogeneous processes, N/IWC should also be higher for temperatures less than -40°C if homogeneous freezing is active. It can be seen in Fig. 2b that N/IWC values can be described as two groups; one for $T < -40^{\circ}\text{C}$ and one for $T > -40^{\circ}\text{C}$. The horizontal bars indicate the range of temperature values in each group while the vertical bars indicate the standard deviation for each group of points. The intersection point gives the mean ratio value for each group. Figure 3 shows \bar{D} for the fresh anvil cirrus related to temperature. Like Fig. 2b, the \bar{D} values cluster into two groups: one for $T < -40^{\circ}\text{C}$ and one for $T > -40^{\circ}\text{C}$. The horizontal and vertical bars follow the same convention as Fig. 2b. Higher nucleation rates should produce smaller values of \bar{D} .

Figures 1, 2b and 3 collectively suggest that a change in ice nucleation mechanism occurs near -40°C when updrafts are sufficiently high (corresponding to fresh anvil cirrus). Figure 1 shows that it is the concentration of the smaller more recently nucleated ice particles in fresh anvils that increase at temperatures below -40°C . For cloud updrafts between 1 and 4 m s^{-1} (common for marine deep convection at cirrus levels), homogeneous freezing predicts N between ~ 1 and 10 cm^{-3} (or 1000 to $10,00 \text{ liter}^{-1}$) for relatively low aerosol particle concentrations ($< \sim 150 \text{ cm}^{-3}$) between -40 and -50°C (Barahona and Nenes 2008). Most of the N values for fresh anvil cirrus in Fig. 2a fall into this approximate range of expected values. The close grouping of N/IWC values in Fig. 2b for $T < -40^{\circ}\text{C}$ indicates that the two N values $\sim 100 \text{ liter}^{-1}$ in Fig. 2a are associated with lower IWCs and perhaps lower updrafts and thus still consistent with homogeneous freezing nucleation. Moreover, entrainment of drier air into the anvils with subsequent sublimation, inhomogeneous mixing and dilution will reduce N . Overall, this analysis indicates that homogeneous freezing appears to be an important process, perhaps the main nucleation process, for fresh anvil cirrus clouds. Therefore RH_i levels should be more than 140% in the region where anvil ice crystals are formed.

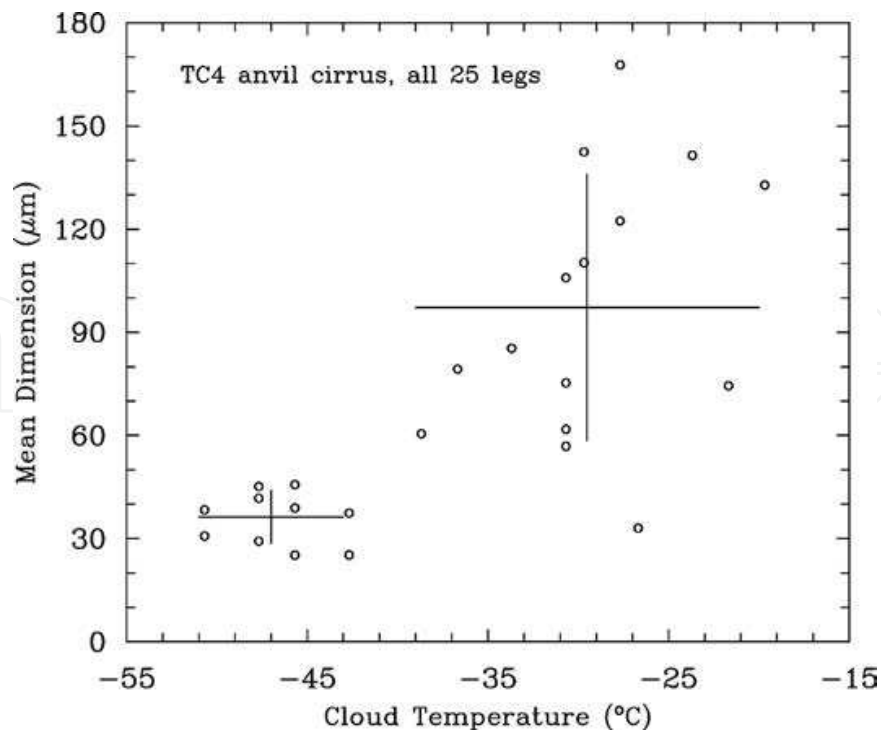


Fig. 3. Mean maximum dimension (i.e. mean ice particle size) related to temperature for the fresh anvil PSDs sampled during TC4.

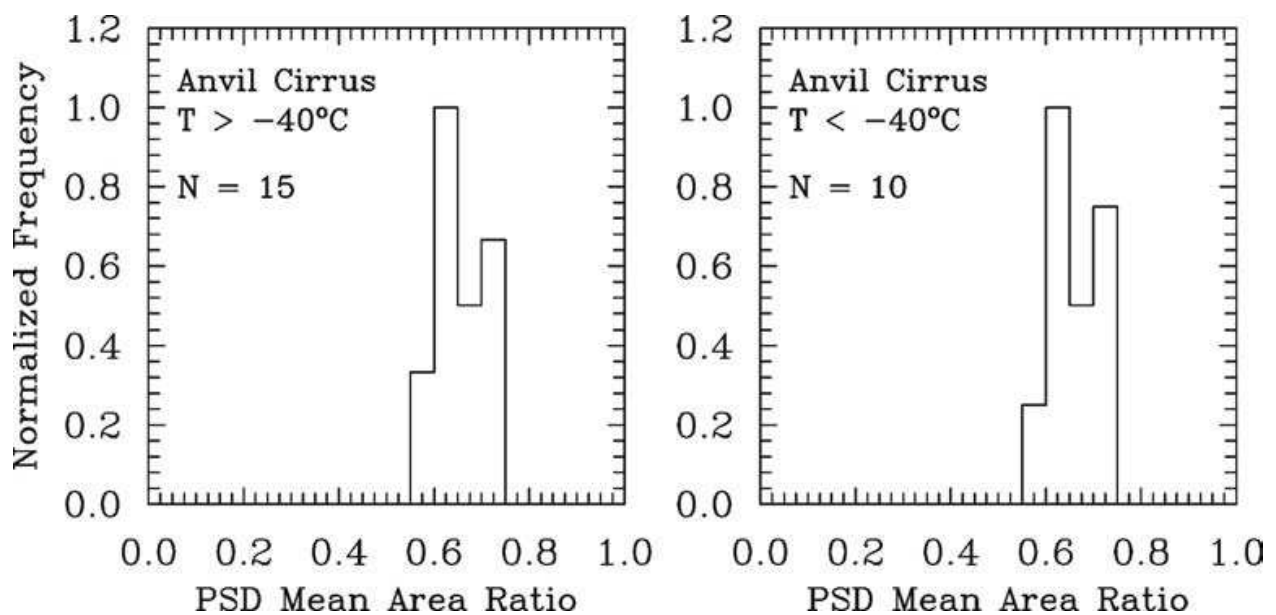


Fig. 4. Normalized frequency distributions of the PSD area ratio for fresh anvil cirrus in two temperature regimes. N refers to the number of PSD sampled.

3.4.1 Temperature dependence of ice particle shape

As mentioned, a change in ice nucleation mechanism may be accompanied by a change in ice crystal habit (i.e. shape). A change in ice particle shape can be detected through evaluating the area ratio (AR) as described in section 3.3. Figure 4 shows the temperature

dependence of the AR for fresh anvil cirrus for temperatures more than -40°C and for temperatures less than -40°C . Since there is no significant change in the normalized frequency distribution between temperature regimes, there is no evidence for a change in ice particle shape. Similar results were obtained for aged anvil and in situ tropical cirrus, although aged anvil cirrus exhibited a frequency value 0.5 in the largest AR bin (corresponding to unity) for $T < -40^{\circ}\text{C}$ (this was not present for $T > -40^{\circ}\text{C}$).

For the fresh anvil cirrus case, the results might be explained if the geometry of the ice nuclei (e.g. irregular mineral dust particle vs. spherical haze or cloud droplets) influences subsequent ice crystal growth patterns. It is reasonable to expect the freezing of supercooled cloud droplets (e.g. via immersion freezing nucleation or contact nucleation (Pruppacher and Klett 2010)) to contribute substantially to new ice production for temperatures greater than -38°C . These same supercooled cloud droplets may experience flash freezing at the -38°C level where all liquid water freezes via homogeneous freezing, as observed by Rosenfeld and Woodley (2000) in strong convective updrafts. This would increase N above the -38°C level but may not result in a change in ice crystal shape.

3.4.2 Application to climate engineering

The above analysis indicates that fresh anvil cirrus may be susceptible to modification by seeding aerosol. But due to the formation mechanism of anvil cirrus, it is less clear just how seeding could be accomplished, since anvil cirrus represents the outflow of deep convection where boundary layer air (typically within 1 km of the surface) has been vertically advected into the anvil cirrus. Thus if the seeding aerosol were dispersed at anvil cirrus levels, and ice crystal production occurs in the convective updraft, there may be little opportunity for the seeding aerosol to produce new ice crystals (scavenging by wet deposition processes could remove most of the aerosol before it sediments to boundary layer levels). Alternatively, the boundary layer could be seeded, but some portion of the seeding aerosol would be scavenged and removed through wet deposition processes in the convective updraft. Fridlind et al. (2004) present evidence indicating mid-tropospheric aerosols contribute substantially to ice nucleation in anvil cirrus, and if this is true, then seeding at temperatures less than -45°C may be effective. These factors make the seeding of tropical anvil cirrus a more complicated proposition relative to synoptic cirrus clouds that are commonly found in the subtropics, the mid-latitudes and the polar regions. Synoptic cirrus clouds are formed in gradually ascending air masses such as lifting motions due to a warm front. Thus seeding the region of atmosphere where they typically form under clear conditions could modify these cirrus clouds once they form in the treated air mass.

To counteract the worst effects of global warming, it may not be necessary to seed anvil cirrus clouds. Both observational evidence and GCM predictions show that global warming due to GHG emissions will affect the polar regions much more than the tropics (Soloman et al., 2007), thus reducing the temperature gradient between the poles and tropics. To counteract this effect, seeding could be applied to only the mid-latitudes and polar regions, increasing OLR from those regions. This would cool the planet where it needs it most, and also act to restore the "normal" temperature gradient between the tropics and polar regions.

3.5 Synoptic cirrus clouds: SPARTICUS results

Synoptic cirrus clouds are characterized by ice formation conditions distinctly different from those of anvil cirrus; updrafts are much weaker (typically $\sim 10\text{-}30\text{ cm s}^{-1}$) and heterogeneous processes like deposition and condensation freezing nucleation may play a larger role. The

SPARTICUS field campaign, designed in part to evaluate the concentrations of small ice crystals in cirrus clouds using improved instruments much less vulnerable to artifacts from ice particle shattering, was conducted over the continental USA, especially over the Southern Great Plains ARM site. Although both anvil and synoptic cirrus clouds were sampled during SPARTICUS, this study considers only the synoptic cirrus clouds. The cirrus clouds were identified as synoptic based on flight notes and satellite observations. There were 13 synoptic cirrus clouds analyzed that met the criteria established for good data quality. This resulted in 174 PSD that were grouped into 5°C temperature intervals and mean PSD were calculated for each temperature interval, producing 11 mean PSD. The number of PSD found in each temperature interval is shown in Fig. 5.

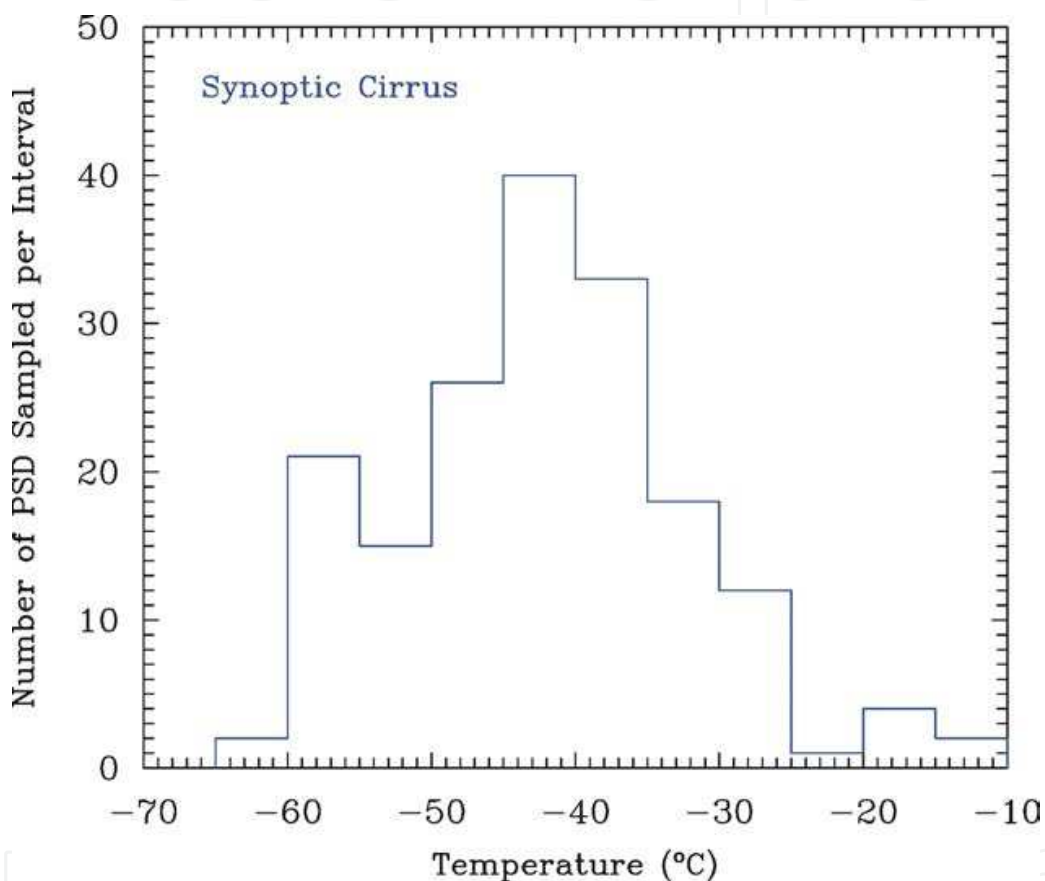


Fig. 5. PSD sampling statistics for SPARTICUS synoptic cirrus, based on 13 cirrus flights during 9 days.

The PSD data processing criteria established for good data quality are as follows: A sampled flight segment begins when the extinction coefficient β_{ext} (twice the measured particle projected area) exceeds 0.1 km^{-1} over a 5 second period. The mean value of β_{ext} and the median mass dimension D_m are calculated were the sample time resolution is 1 second (i.e. 1 Hz). If the maximum values of β_{ext} and D_m do not exceed their mean value by a factor of two, and their minimum values are not less than 0.4 times the mean value, then the process continues. Sampling time is then increased in 1-second increments; every second the value of β_{ext} and D_m are calculated and compared against their respective cumulative mean values. If they do not exceed two times the cumulative mean and are not less than 0.4 times the cumulative mean, then the segment passes and another second is added. A segment must

reach 60 seconds in length to be kept and when an acceptable segment reaches 120 seconds in length the first half is cut off and kept as a PSD measurement and the second half continues adding seconds provided the above criteria are met. The process goes on in this way such that all PSD measurements have sampling times between 60 and 120 seconds. The method makes use of a large portion of the data while insuring relatively stable cloud conditions.

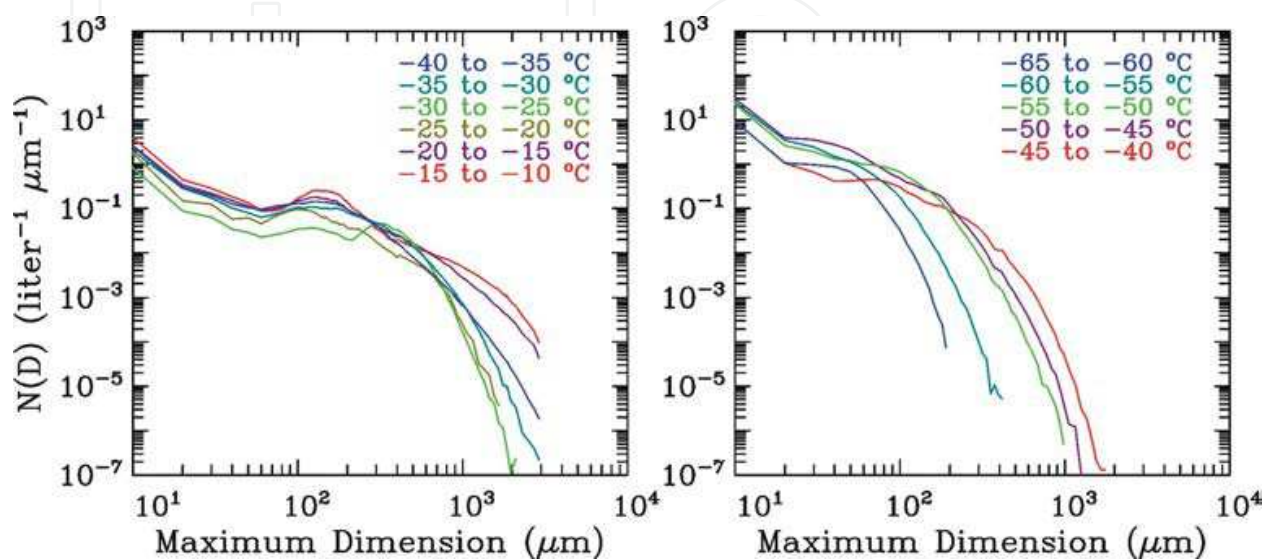


Fig. 6. Temperature dependence of the synoptic PSD during SPARTICUS.

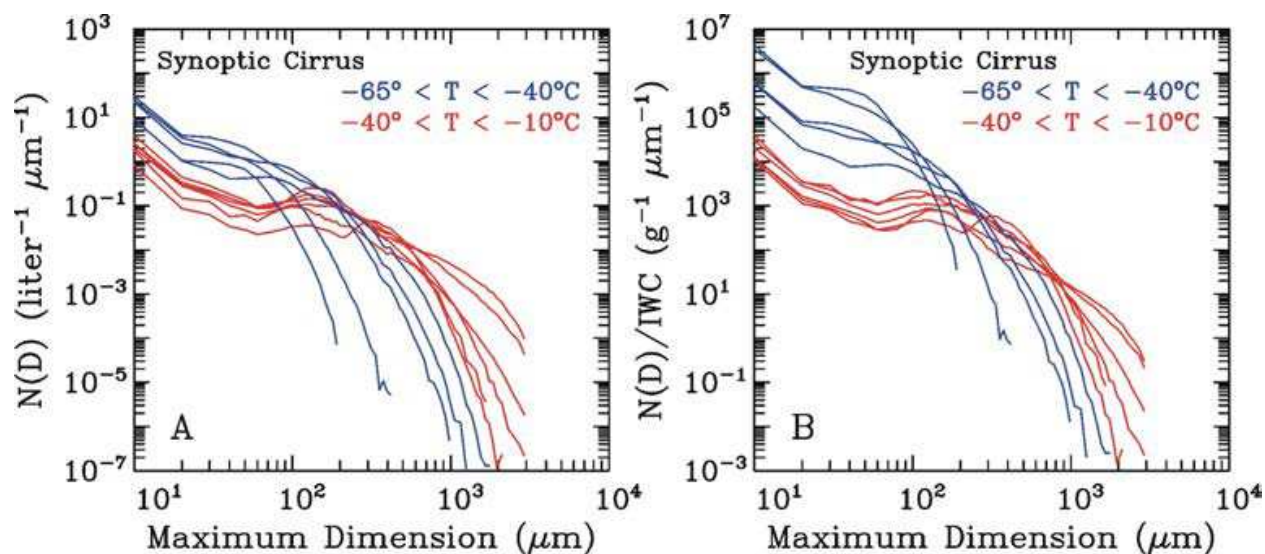


Fig. 7: A: As in Fig. 6 but with all 11 mean PSD shown together and color-coded by temperature regime. B: The same PSD as in panel A, but divided by their corresponding IWC to express the impact of ice nucleation rates on PSD.

The temperature dependence of the PSD that were processed and selected in this way is shown in Fig. 6, with PSD warmer than -40°C on the left and PSD colder than -40°C on the right. A clear difference in PSD shape is evident, with the warmer PSD being bimodal and the colder PSD being approximately mono-modal. As with the anvil cirrus, this could be

explained by differences in ice crystal nucleation rates, with higher rates regarding the mono-modal PSD. Lower nucleation rates would change the balance between small ice crystal production rates and small ice crystal removal rates from aggregation, perhaps resulting in bimodal PSD as shown in Fig. 6 and discussed in Section 3.4.

The mean PSD are again shown in Fig. 7a, but are color-coded to show the transition in PSD shape at -40°C . A transition in ice nucleation rates at -40°C is better expressed by dividing each PSD by its corresponding IWC, as shown in Fig. 7b, showing the number of ice particles per gram of condensate per μm size interval. Differences in nucleation rates are now easily recognized by the concentration differences of the smaller ice crystals. This is similar in principle to the N/IWC ratio as described in relation to Fig. 2b. Figures 6 and 7 demonstrate that at -40°C , there are distinct changes in PSD shape, and for small ice crystals ($D < 100 \mu\text{m}$), distinct changes in N and N/IWC .

The total number concentration N and the N/IWC ratio are plotted for the mean PSD, as shown in Fig. 8. When less than three PSD occurred in a given temperature interval, no standard deviation was calculated (see Fig. 5 for sampling statistics). Merely relating N to PSD temperature may not reveal a change in nucleation rate unambiguously, but the N/IWC ratio reveals a change in nucleation rate near -40°C more clearly. As predicted by homogeneous freezing nucleation theory (Barahona and Nenes 2008), J should be most sensitive to changes in temperature and cloud updraft (RH_i). The increase in N/IWC with colder temperatures ($< -40^{\circ}\text{C}$) appears consistent with homogeneous freezing theory. Moreover, the N values shown in Fig. 8a also appear consistent with those predicted for updrafts typical of synoptic cirrus clouds ($10\text{-}30 \text{ cm s}^{-1}$) at temperatures between -40 and -60°C and for an aerosol particle concentration of 200 cm^{-3} , where the vapor deposition coefficient for ice growth ranges from 0.1 to 1.0 (Barahona and Nenes 2008). However, continental aerosol concentrations measured over Florida between 5 km and 15 km ranged from 3000 cm^{-3} to 100 cm^{-3} (Fridlind et al. 2004). If typical aerosol concentrations during SPARTICUS were on the order of $\sim 1000 \text{ cm}^{-3}$ at cold cirrus levels, then the observed N s may be explained by entrainment and dilution processes.

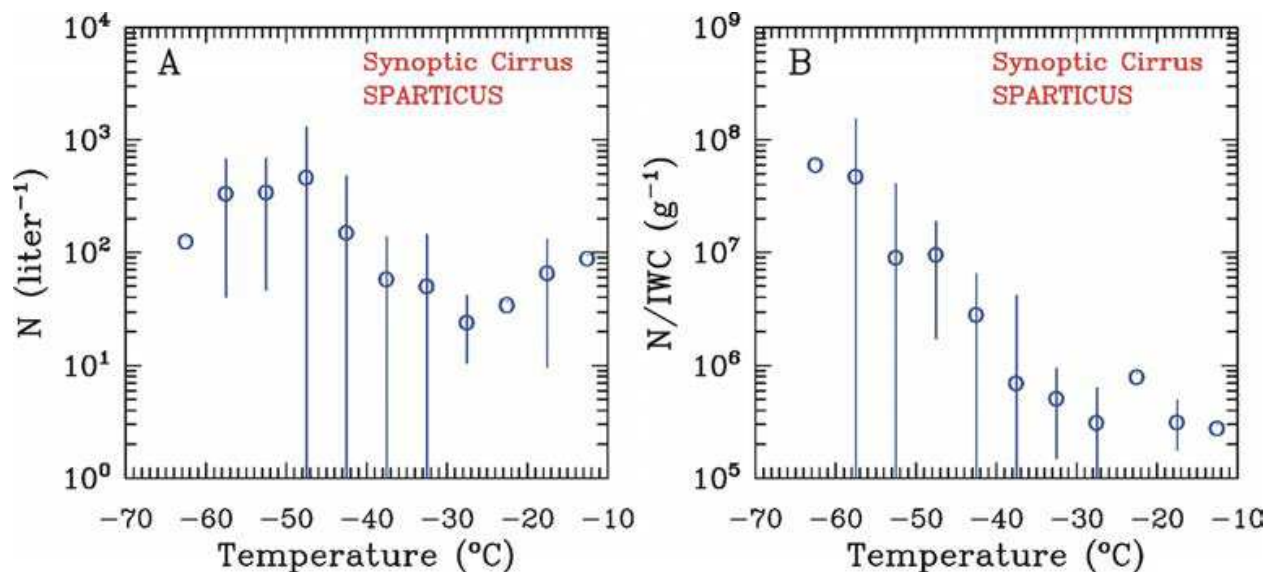


Fig. 8. Temperature dependence of the total ice particle number concentration and the N/IWC ratio based on mean PSDs. Vertical bars are standard deviations. Data points based on less than 3 PSD have no standard deviation.

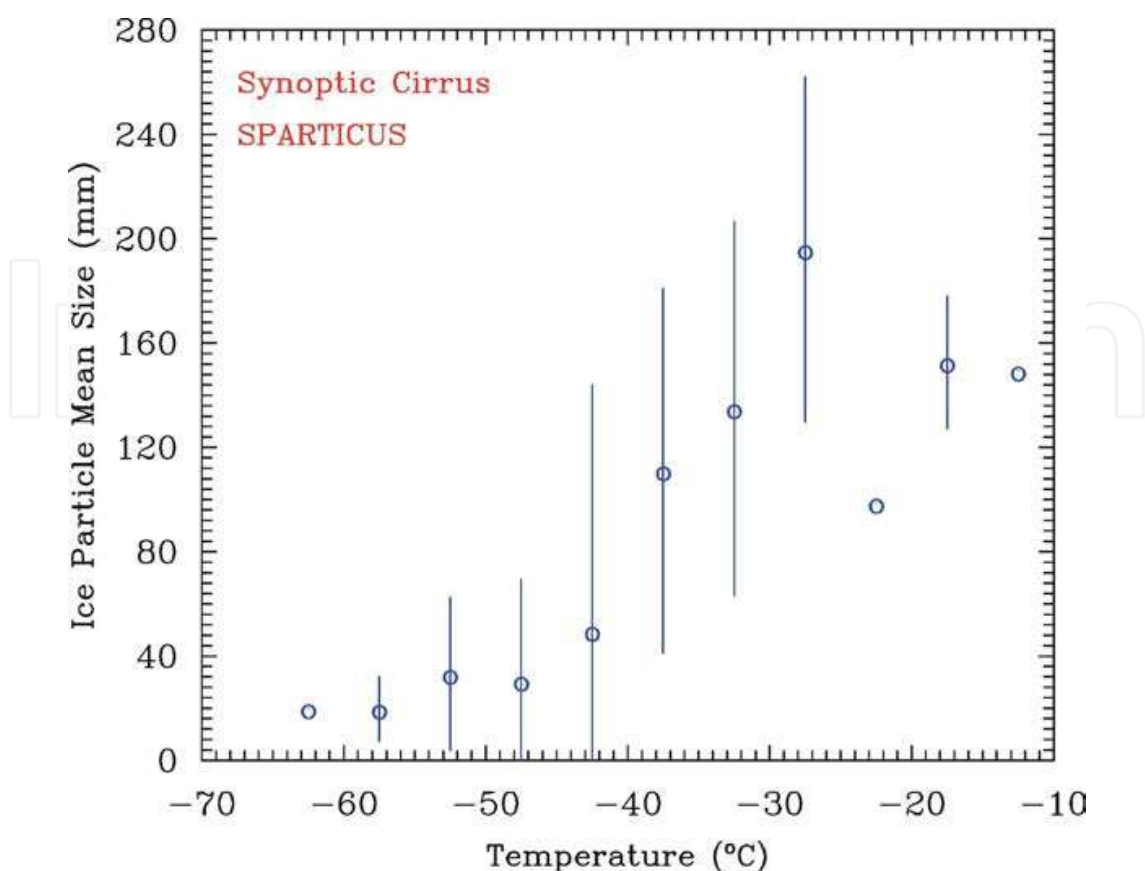


Fig. 9. The temperature dependence of the mean maximum dimension from the mean SPARTICUS PSDs. Points derived from less than 3 individual PSD have no standard deviation.

The same basic pattern shown by the N/IWC ratio is repeated by plotting the mean size of the mean PSD against temperature, as shown in Fig. 9. As ice nucleation rates increase, the mean ice particle size generally decreases.

3.5.1 Temperature dependence of ice particle shape

As mentioned, a change in nucleation rate could be accompanied by a change in ice crystal habit (i.e. shape), depending upon the ice nucleation mechanisms involved. To explore this possibility, normalized frequency distributions of the mean area ratio of a PSD, for all of the synoptic PSDs processed, were developed for two temperature regimes: more than -40°C and less than -40°C . This is shown in Fig. 10. When the size threshold was changed from $60\ \mu\text{m}$ to $100\ \mu\text{m}$ (i.e. excluding particle sizes less than $100\ \mu\text{m}$ from the analysis), the results were almost the same. This is clearly an unambiguous signal that ice particle shapes generally differ between these temperature regimes. Figure 10 is strikingly different than the similar analysis of area ratios for fresh anvil cirrus shown in Fig. 4, suggesting that the ice growth physics in mid-latitude synoptic cirrus clouds differs appreciably from that of anvil cirrus. The solitary peak near unity for temperatures less than -40°C indicates that these ice crystals are generally either quasi-spherical or isometric (i.e. having nearly equal length and width) in geometry. Since homogeneous freezing nucleation commences at about -38°C , these results strongly suggest that this nucleation mechanism is the reason for the observed change in ice particle morphology. Moreover, they also indicate that almost all of the ice

particles less than -40°C have the same shape. For almost all the ice particles to have the same shape, a common origin may be implied, suggesting that homogeneous freezing produced almost all of the ice particles.

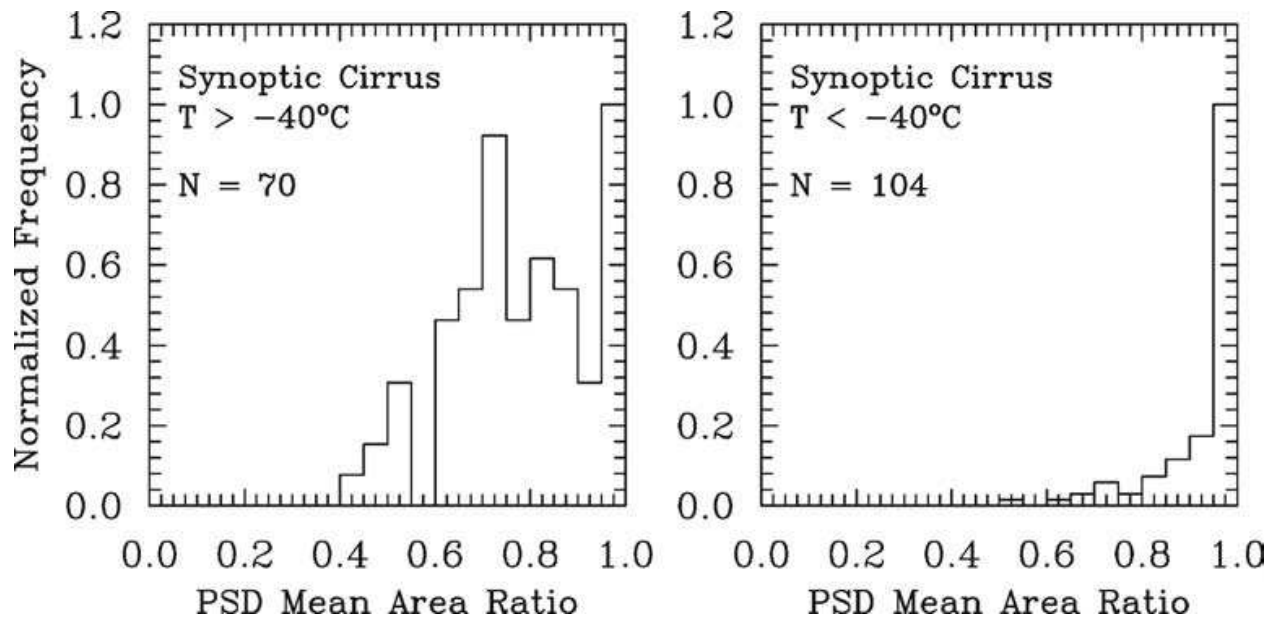


Fig. 10. Normalized probability distribution functions of the mean area ratio for all 174 synoptic cirrus PSDs, divided into two temperature regimes. N refers to the number of PSDs evaluated in each temperature regime.

To evaluate how rapidly this transition in ice particle shape occurs, the PSD area ratio was determined for each of the mean PSD and related to the mean temperature of each interval. This is shown in Fig. 11. The PSD area ratio is lowest at temperatures warmer than -20°C where dendritic ice crystals can grow and ice particles tend to be more aggregated. Between -20 and -40°C , the mean area ratio is virtually flat, but increases abruptly for temperatures less than -40°C . If we attribute this abrupt change near -40°C to homogeneous freezing nucleation, then the onset of homogeneous freezing with decreasing temperature appears rapid.

At temperatures warmer than -40°C , the gradually ascending air motions characterizing synoptic cirrus may make droplet freezing a less likely source of ice crystals since the RH_i levels will be lower with lower updrafts. That is, RH_i may never exceed the water saturation threshold that would allow cloud droplets to form and freeze. This would mean that heterogeneous nucleation processes would more likely occur through deposition nucleation and condensation freezing nucleation rather than droplet-freezing, contact nucleation on cloud droplets and immersion freezing within cloud droplets. If true, the ice crystal shapes produced through nucleation mechanisms not involving cloud droplets might be different than for ice crystals beginning from a frozen cloud droplet. At temperatures colder than -40°C , homogeneous freezing occurs on spherical haze droplets and ice germs begin growing as spheres. On this basis there could be a change in ice crystal shape occurring $\sim -40^{\circ}\text{C}$.

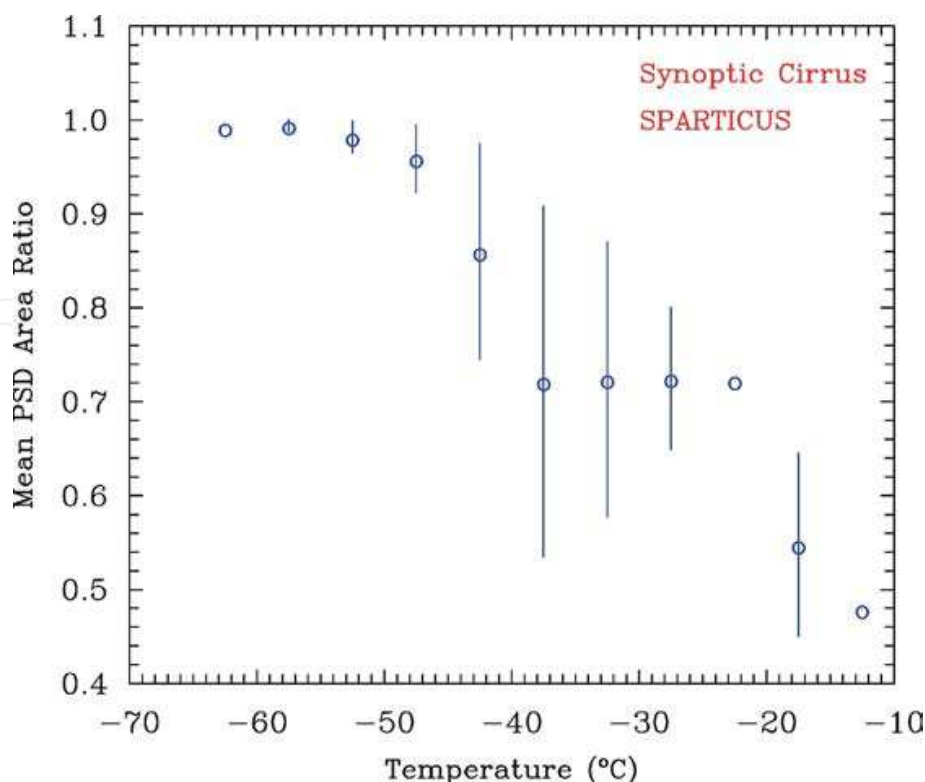


Fig. 11. Temperature dependence of the area ratio calculated from the mean PSD. Vertical bars are standard deviations. Points without such bars have less than 3 PSD samples.

In a dynamically, microphysically and radiatively complex system like the upper troposphere, things are often not what they appear to be. An alternate explanation for the observed ice particle morphology is that when moist layers of the upper troposphere gradually ascend, they become supersaturated and ultimately exceed the threshold RH_i for homogeneous freezing nucleation. Relatively high concentrations of ice crystals are then produced throughout this layer and grow in the supersaturated environment, rapidly drawing down RH_i levels close to ice saturation. The chance of an aircraft sampling regions near the homogeneous freezing threshold would be relatively small since such regions would be transient and short-lived. This appears consistent with the extensive observations in Kramer et al. (2009), who found similar maximum RH_i levels in both clear-sky and inside-cirrus conditions, with maximum RH_i near the homogeneous freezing threshold. For inside-cirrus conditions between -33 and -90°C , RH_i was most commonly found between 85% and 115%. Under such conditions, ice will either grow slowly or slowly sublimate. Ice crystals grown in the laboratory between -40° and -70°C at RH_i between 101% and 110% exhibit isometric growth tendencies, with short columns, thick plates and irregular compact crystals common between -20° and -70°C (Bailey and Hallett 2004, 2009). This is shown in Fig. 12, which is an adaptation of Fig. 5 in Bailey and Hallett (2009), courtesy of Dr. Matthew Bailey. This suggests that the results in Fig. 10 and 11 at temperatures less than -40°C might be due to a low RH_i growth environment. The Krämer et al. observations also suggest that heterogeneous ice nucleation processes may not always be active for RH_i well above ice saturation since peak RH_i 's were similar for both clear-sky and inside-cirrus conditions. Overall, this explanation attempts to broach the paradox of high RH_i required for homogeneous nucleation and low RH_i commonly found in cirrus clouds and associated with ice particle shapes at temperatures less than -40°C . This description of cirrus cloud

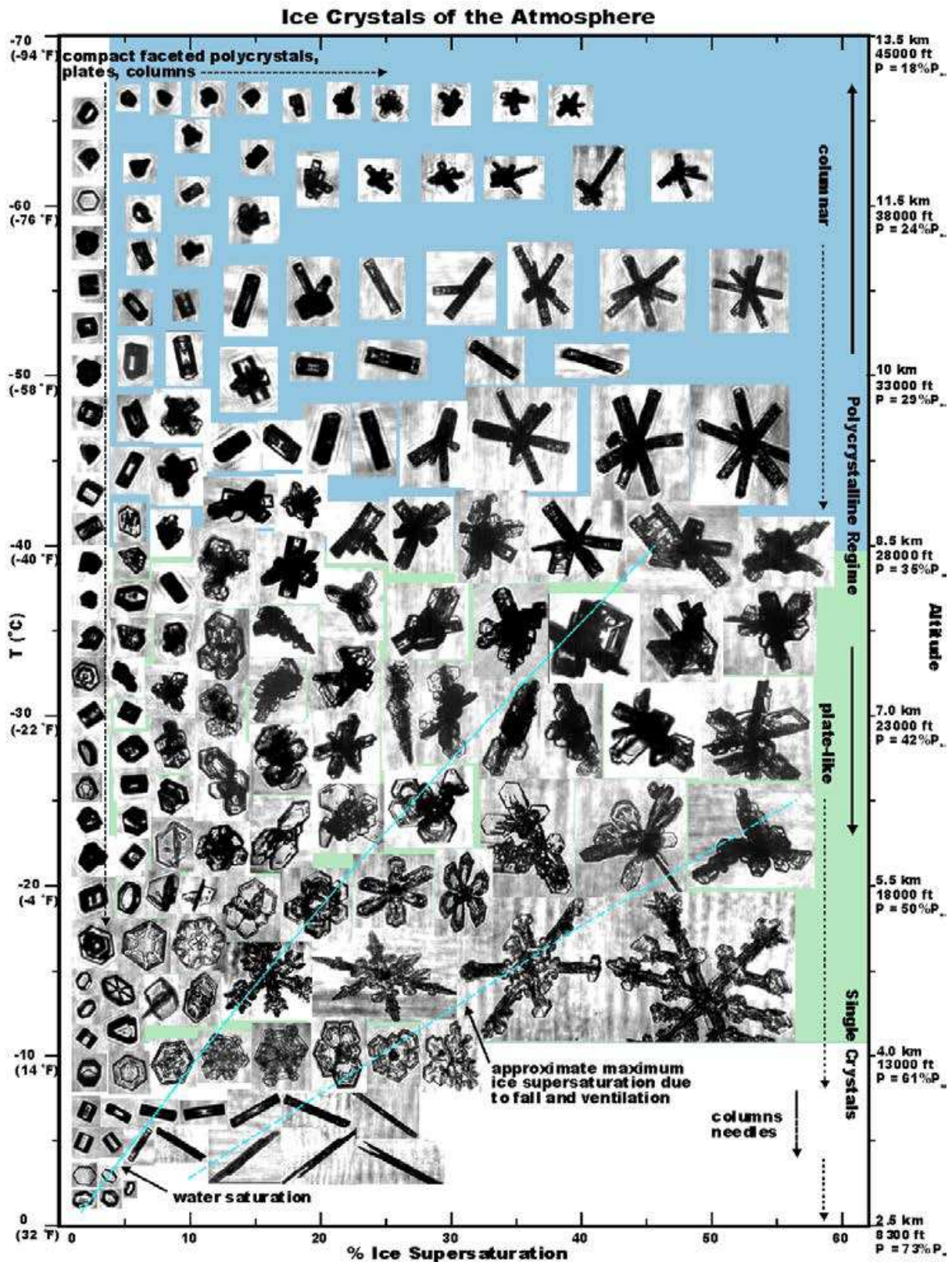


Fig. 12. Ice crystal shape as a function of formation temperature and supersaturation with respect to ice. Right axis gives approximate height and pressure of crystal formation where P_0 is standard atmosphere pressure. Figure adapted from Bailey and Hallett (2009), courtesy of Dr. Bailey.

formation is similar to that of others (e.g. Haag et al. 2003; Spichtinger et al. 2004) except it offers an explanation of ice particle shape. It may not apply in cases where cirrus emanate from a convective “seeder” region, such as cirrus uncinus, where ice crystals nucleate and grow in more convective air, resulting in fall streaks as these ice crystals descend from this region. Rather, in this description, ice is produced throughout the whole moist layer without a specific “source region” involved, which appears consistent with most visual observations of cirrus clouds. In summary, two hypotheses have been described for explaining the change in ice crystal shape; one based on the morphology of the original ice germ and another based on the average supersaturation level within the cirrus cloud. These need not be mutually exclusive, and both processes could be relevant in determining ice particle shape.

Similar observations are reported in Lawson et al. (2006b), where extensive measurements in mid-latitude cirrus clouds were made using the Cloud Particle Imager (CPI) probe. The CPI images ice particles with a resolution of 3 μm , yielding very detailed images of their structure (Lawson et al. 2001). The CPI analysis in Lawson et al. (2006b) showed that for temperatures warmer than -40°C and sizes greater than 50 μm in length, number concentrations were dominated by rosette-shaped ice crystals ($\sim 57\%$), whereas at colder temperatures, irregular ice crystals dominated, increasing in abundance with decreasing temperature. The irregular crystal category in Lawson et al. (2006b) corresponds to ice particle shapes that do not fit into “traditional” shape categories such as hexagonal columns and plates, bullet rosettes, aggregates, etc. These irregular ice crystals were generally isometric, having “blocky” and sometimes quasi-spherical shapes. Examples of ice crystals in mid-latitude cirrus clouds, including these blocky and bullet rosette ice crystals that appear to dominate the homogeneous freezing and heterogeneous nucleation zones, respectively, are shown in Fig. 13.

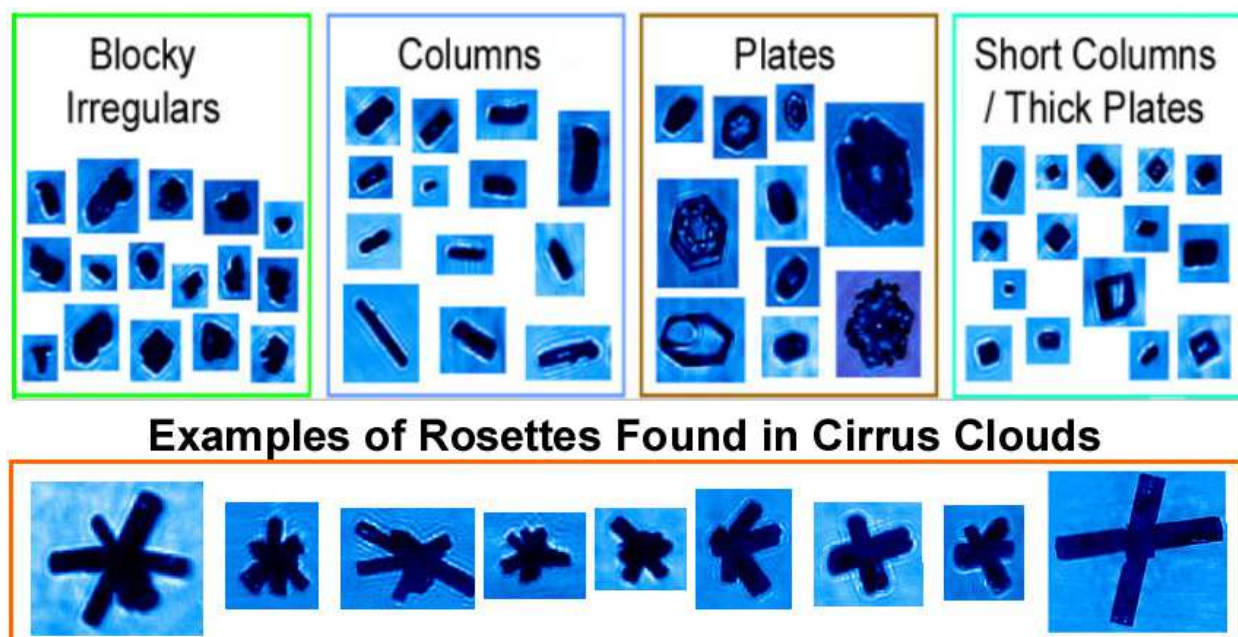


Fig. 13. CPI images of ice crystals sampled in mid-latitude cirrus clouds, with blocky irregulars being most common at temperatures below -40°C and rosette-shaped crystals being more common at warmer temperatures.

3.5.2 Temperature dependence of the ice fall-speed

As discussed in Section 2, the ice fall-speed has a strong impact on climate sensitivity in some GCMs. It is therefore critical to understand the processes determining the ice fall-speed, such as ice nucleation and ice particle morphology. The way these processes conspire to determine the mass-weighted ice fall-speed V_m is shown in Fig. 14. While V_m is roughly constant between -20° and -40°C , V_m abruptly decreases at colder temperatures. Ice particle shape impacts V_m through the ice particle mass/area ratio, with higher ratios increasing V_m . These results illustrate how changes in ice particle size (a product of nucleation rates) have a greater impact on V_m than ice particle shape does, since the mass/area ratio should be greater for compact, high density irregular ice particles relative to side planes and rosette-shaped crystals common to temperatures warmer than -40°C (Bailey and Hallett, 2009; Lawson et al. 2006b). Thus it appears that the onset of homogeneous freezing nucleation produces a rather dramatic decrease in V_m in spite of an expected increase in the ice particle mass/area ratio. As discussed in Section 2, this should increase the lifetime and cloud coverage of cirrus at temperatures below -40°C relative to warmer cirrus clouds. In addition, these results indicate that V_m might be substantially increased by seeding aerosol for temperatures colder than about -45°C . This would reduce cirrus coverage and increase OLR, cooling the planet.

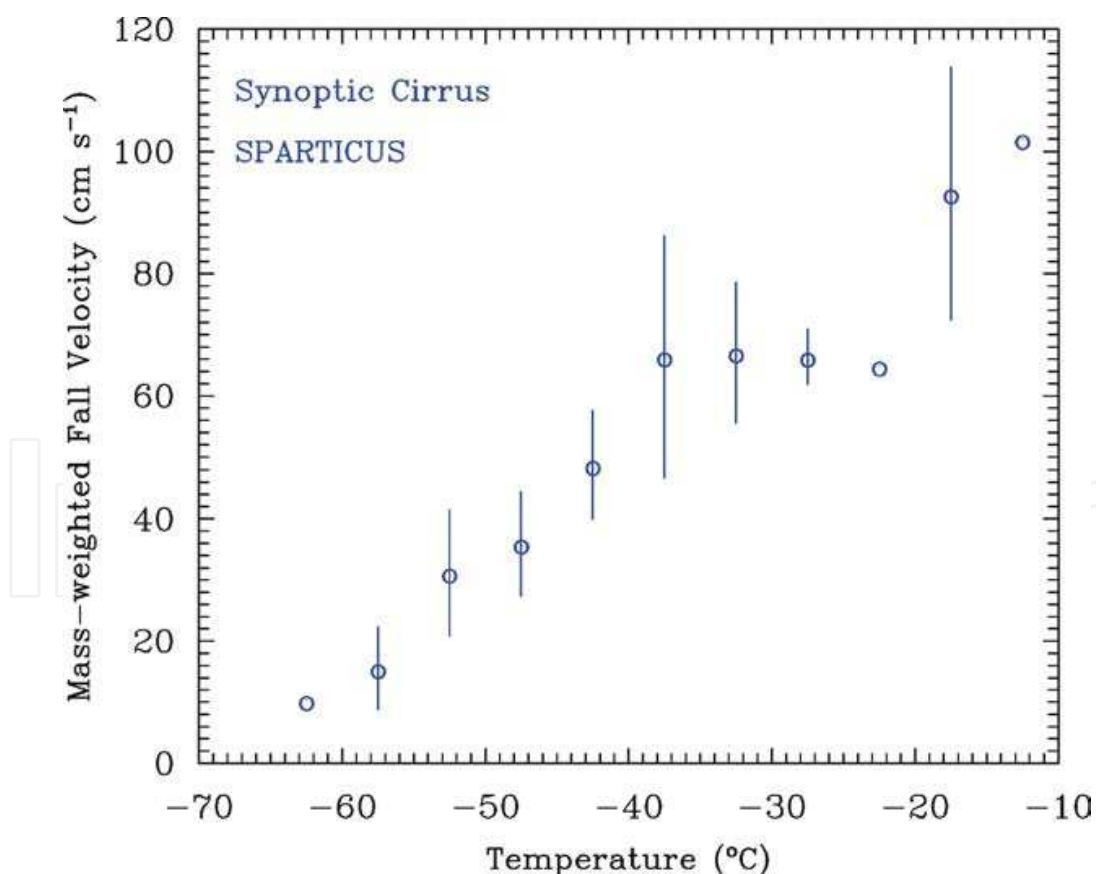


Fig. 14. Temperature dependence of V_m corresponding to the mean PSD. Vertical bars indicate standard deviations. Data points without bars are based on fewer than 3 PSD samples.

Similar V_m results were not found for tropical anvil cirrus clouds, and the temperature dependence of V_m for these clouds is reported in Mitchell et al. (2011). The PSD in these clouds for temperatures less than -40°C were generally characterized by relatively high IWCs, indicating strong updrafts and condensation rates. As a result, the concentrations of larger ice particles in these PSD were relatively high, increasing V_m , and a significant decrease in V_m at temperatures below -40°C was not observed. However, only 25 PSD were sampled for fresh anvil cirrus, and a larger dataset could reveal a temperature dependence for V_m similar to mid-latitude synoptic cirrus.

4. Over-cooling the planet

In section 2.4 a GCM study by Lohmann et al. (2008) was discussed that demonstrated that a substantial global cooling effect might be possible through seeding with heterogeneous ice nuclei that activate at $\sim \text{RH}_i = 130\%$. This produced an 11% increase in effective ice particle size and a corresponding increase in the ice fall-speed, which resulted in a mean global net cooling of $\sim 2.7 \text{ W m}^{-2}$. This compares with a net radiative forcing due to a CO_2 doubling of 3.7 W m^{-2} . Since we are suggesting the use of very efficient ice nuclei having RH_i 's $\sim 105\%$ or lower, the impact of cirrus seeding on the cirrus cloud ice fall-speed V_m and hence global cooling could be considerably greater than in the Lohmann et al. study. This presents the possibility of "over-cooling" the planet.

As mentioned, cirrus cloud climate engineering could be accomplished by only seeding cirrus clouds outside the tropics. As temperatures in the mid-latitudes and polar regions decrease, snow-cover in those regions would likely increase, with increases in Arctic sea-ice and polar ice sheets. This would reflect more sunlight to space. The climate system is very sensitive to these climate feedback effects (Thompson and Sieber, 2010), which could substantially amplify the direct radiative cooling produced by the cirrus cloud climate engineering.

Since currently the treatment of cirrus clouds and climate feedbacks in GCMs can vary considerably, the cooling effect predicted by different GCMs could vary substantially regarding this climate engineering approach. Thus it may be difficult to determine at this time through GCM simulations how much and how rapidly the planet would cool, although a range of estimates would be useful. To know whether the climate is responding at an appropriate rate, it will be necessary to closely monitor Earth's radiation budget and the global climate.

GCM simulations indicate there is considerable time lag (e.g. decades) for a change in globally averaged net radiative forcing to fully manifest a corresponding change in global mean temperature. Thus temperature is not an appropriate variable to use for monitoring the effects of climate engineering. Rather shortwave and longwave radiative forcing should be monitored, as these will respond immediately to changes in the climate system (Evans 2010). This should involve both satellite and ground-based monitoring networks (Evans 2010).

5. Social, ethical and political implications of climate engineering

Global warming is just one of many challenges facing humanity, but it is unique in that it could produce a severe and irreversible change in the global climate if the global mean surface temperature exceeds 2°C (relative to the beginning of the industrial era) and trigger key tipping points in the climate system, such as the melting of the polar ice caps (Hansen et al., 2008). Whether humankind could survive such a climate change is an open question.

There appears to be a growing consensus among scientists that, while no one wants to geo-engineer the climate, some kind of geo-engineering will be needed in combination with severe GHG mitigation to avoid triggering climate system tipping points that would result in catastrophic climate disasters, irreversibly locking the climate into a much warmer state (e.g., Thompson and Sieber, 2010; Arora et al. 2011; Greene et al., 2010; Hansen et al., 2008). And if the Earth's climate sensitivity is 2 to 4 times more than GCMs have predicted based on paleoclimate research (Kiehl 2011), it may already be too late to avoid passing the 2°C threshold.

From another point of view, perhaps our fixation with technological solutions is distracting us from larger ontological questions regarding our relationship to Nature and our spiritual being. What if the root of the problem lies in Man's consciousness and the relentless pursuit of insatiable desires? Can the planet handle this? Is it possible that global warming is Nature's way of getting us "back on track" to live in harmony with Nature and with each other? Perhaps climate engineering would at best merely postpone and then intensify an apocalyptic tragedy if human consciousness is not transformed. Perhaps these are the real issues that climate change is presenting to us. A more extensive exploration of this viewpoint is given in an essay by Prof. Clive Hamilton (2011).

If climate engineering in combination with resource conservation, renewable energy systems and GHG reduction are all needed for our survival, then it behooves us to explore what new opportunities climate engineering presents for manifesting positive social and political changes in the world. Since it would affect the entire world, climate engineering should be internationally organized and executed, requiring the cooperation of all the nations of the world. Seen in this way, global warming may bring about a situation mandating the cooperation of the entire human race, asking people and nations to go beyond their immediate self-interest and act for the good of the whole planet. The future climate of the planet may depend on whether nations can cooperate in a spirit of shared-sacrifice, and for democratic nations, it depends on whether the people themselves can act in this way. As it has always been, our collective destiny depends on our collective consciousness and our ability to transform it to meet the challenges of our time.

6. Summary and conclusions

A new approach to climate engineering, described in Mitchell and Finnegan (2009), has been reviewed in Section 2. A key principle determining the success of this approach is the ability of cloud seeding aerosol to out-compete the natural ice nuclei for water vapor when cirrus cloud temperatures are below -40°C. This vapor competition effect will be most pronounced, with cloud modification due to seeding very likely, if homogeneous freezing nucleation plays an important role in ice crystal production. This chapter provides new findings regarding the importance of homogeneous freezing in cirrus clouds relative to heterogeneous nucleation processes. These findings were made possible through recent improvements in ice particle measuring probes consisting of (1) a dramatic reduction in small ice particle artifacts due to the shattering of natural ice particles at the probe inlet tube, (2) the ability to accurately measure ice crystal concentrations down to 10 µm in size and (3) 10 µm resolution for imaging the shapes of ice crystals, resulting in more accurate ice particle area ratios as described in Section 3.3.

For mid-latitude synoptic cirrus around -40°C, these new measurements show that clear changes in the ice particle size distribution (PSD) and its properties occur regarding (1) PSD

shape, (2) total number concentration-to-ice water content ratio (N/IWC), (3) PSD mean size, (4) PSD mean area ratio and (5) the mass-weighted fall velocity (V_m). These five changes are consistent with a change in ice nucleation mechanism, with heterogeneous nucleation processes active at temperatures warmer than -40°C and homogeneous freezing nucleation at temperatures colder than -40°C . The change in V_m implies that synoptic cirrus colder than -40°C will have longer lifetimes and greater cloud coverage than warmer cirrus clouds, all other relevant factors remaining equal.

Regarding tropical anvil cirrus clouds near -40°C , changes were observed in the first 3 of the 5 attributes listed above. For these fresh anvil cirrus clouds, homogeneous freezing nucleation also appears to be an important process for ice crystal production. The lack of a change in ice particle shape at -40°C may be due different dominant heterogeneous ice nucleation processes (e.g. droplet freezing) and less of a change in RH_i between temperature regimes. The lack of a change in V_m behavior near -40°C may be due to relatively high condensation rates associated with PSDs sampled at temperatures less than -40°C , producing higher concentrations of both small and larger ice particles. Note that V_m depends mostly on the larger ice particle concentrations (Mitchell et al. 2011). A larger PSD dataset for tropical anvil cirrus would help determine whether this result was general or not.

In regards to synoptic cirrus clouds, we feel that the evidence for homogeneous freezing nucleation being a major source of ice crystals is exceptionally strong. While it is entirely possible that heterogeneous processes are also an important source of ice crystals at temperatures below -40°C , the dramatic change in ice particle shape near -40°C suggests that the vast majority of ice crystals came from homogeneous freezing. These ice crystals below -40°C had area ratios near unity, indicating compact-irregular, isometric and/or quasi-spherical ice crystal shapes. Such ice crystals are associated with growth conditions having low RH_i ($< 110\%$), presenting the following paradox: how is it possible for ice crystal growth below -40°C to correspond to low RH_i while these same ice crystals were formed at high RH_i needed for homogeneous nucleation? This was explained by postulating that moist layers in the upper troposphere experience the RH_i threshold for homogeneous freezing nucleation, which then produces sufficiently high concentrations of ice crystals to vigorously compete for water vapor immediately after nucleation. This rapidly lowers the RH_i throughout the layer, creating a low RH_i environment for subsequent growth. Heterogeneous processes can still contribute new ice crystals at these lower RH_i but at lower rates. This explanation is similar to that proposed by Haag et al. (2003) and others, and appears consistent with the RH_i measurements in Krämer et al. (2009), noting that high RH_i regions corresponding to homogeneous nucleation should be transient and short-lived, making it unlikely that research aircraft would sample such regions. Overall, these results indicate that synoptic cirrus clouds could be modified relatively easily through seeding aerosol in the right concentration range to increase V_m , which should decrease cirrus cloud cover, ice water path and optical depth, and thus increase outgoing longwave radiation (OLR).

Due to the nature of global warming, with surface temperatures increasing in the polar regions and mid-latitudes considerably more than in equatorial regions, it makes sense to enhance OLR primarily in the mid-latitudes and polar regions. This would also stabilize the temperature gradient between the equatorial and polar regions and thus limit changes to the storm track (i.e. jet stream) that might otherwise be altered due to global warming.

Although the seeding of cirrus clouds to reduce global warming is referred to as a climate engineering approach, this is somewhat of a misnomer since "engineering" generally refers

to the application of well established scientific principles. In the case of cirrus cloud engineering, the science is still in the process of being worked out. Moreover, the scientific issues involved are not only relevant to climate engineering; they are of fundamental importance to our understanding of cirrus clouds, their role in the climate system, and their representation in climate models. Therefore this research should also be viewed in the broader context of basic research on the climate system, having the potential to improve the ability of climate models to predict future climates.

GCMs used to study this climate engineering approach should have a physically realistic treatment of cirrus clouds, including a realistic cirrus cloud coverage and treatment of supersaturation with respect to ice. Moreover, realistic representations of climate feedback effects are needed, such as those associated with Arctic sea-ice and polar ice sheets. The latter is needed since the direct radiative cooling from cirrus cloud modification may enhance these feedbacks in the climate system and thus amplify the direct cooling.

While the social and political ramifications of climate engineering are many, its potential role as a catalyst for increased global cooperation should not be discounted. It appears that cirrus cloud climate engineering would be relatively inexpensive to implement.

While this climate engineering approach may appear promising, it is absolutely critical to understand that this is not a “silver bullet” for the global warming problem. To think in these terms would be a prescription for sudden, catastrophic climate change, since there is only so much “wiggle room” in the climate system. That is, a successful modification of cirrus clouds at temperatures below -40°C might cool the planet for a limited period of time, but not indefinitely, and at current GHG emission rates it would not take long for any cirrus OLR benefit to be neutralized. A “business as usual” GHG emission scenario would result in a sudden and dramatic increase in global temperatures once any cirrus OLR benefit was exhausted, and such a temperature increase might make human life on Earth unbearable. Therefore this cirrus climate engineering method may “buy time” for societies to transition to renewable non-carbon fuels and to implement carbon sequestration methods that address ocean acidification, but it should never be viewed as a solution to global warming.

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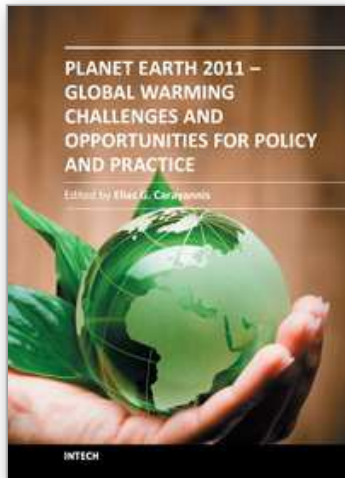
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The failure of the UN climate change summit in Copenhagen in December 2009 to effectively reach a global agreement on emission reduction targets, led many within the developing world to view this as a reversal of the Kyoto Protocol and an attempt by the developed nations to shirk out of their responsibility for climate change. The issue of global warming has been at the top of the political agenda for a number of years and has become even more pressing with the rapid industrialization taking place in China and India. This book looks at the effects of climate change throughout different regions of the world and discusses to what extent cleantech and environmental initiatives such as the destruction of fluorinated greenhouse gases, biofuels, and the role of plant breeding and biotechnology. The book concludes with an insight into the socio-religious impact that global warming has, citing Christianity and Islam.

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