The Origin of High Ice Crystal Number Densities in Cirrus Clouds

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ABSTRACT

Recent measurements with four independent particle instruments in cirrus clouds, which formed without convective or orographic influence, report high number densities of ice particles (as high as \( n_{\text{ice}} = 50 \text{ cm}^{-3} \)) embedded in broad density distributions (\( n_{\text{ice}} = 0.1-50 \text{ cm}^{-3} \)). It is shown here that small-scale temperature fluctuations related to gravity waves, mechanical turbulence, or other small-scale air motions are required to explain these observations. These waves have typical peak-to-peak amplitudes of 1–2 K and frequencies of up to \( 10 \text{ h}^{-1} \), corresponding to instantaneous cooling rates of up to 60 K \( \text{ h}^{-1} \). Such waves remain unresolved in even the most advanced state-of-the-art global atmospheric models. Given the ubiquitous nature of these fluctuations, it is suggested that the character of young in situ forming cirrus clouds is mostly determined by homogeneous freezing of ice in solution droplets, driven by a broad range of small-scale fluctuations (period \( \approx \) a few minutes) with moderate to high cooling rates (1–100 K \( \text{ h}^{-1} \)).

1. Introduction

Cirrus clouds cover more than 20% of the planet (Lin et al. 1996) and play an important role in determining the earth’s climate. Besides cloud altitude and thickness, the microphysical characteristics, such as number density, size, and shape, of the ice crystals also have a large influence on the radiative properties of the clouds (Smith et al. 1998). The size of the crystals determines their sedimentation velocity and therefore the cloud lifetime and the humidity of the upper troposphere, and also the humidity of the stratosphere, as this is partly controlled by the dehydration potential of cirrus clouds (Jensen et al. 2001). If the total available water is distributed among a few large particles, it readily sediments and the air is dried more efficiently than if there is a higher number density of smaller particles. Chemical reactions occurring on ice cloud particles are influenced by the available surface area of ice, which again depends on the size and number density of the particles. At a given relative humidity, the size that the ice particles can finally reach depends only on the number density of ice particles nucleated. Therefore the radiative and microphysical properties of a cloud are strongly influenced by the number density. Thus it is vital that the nucleation process is correctly parameterized in climate models.

Measurements of the number density of ice particles in cirrus clouds are often of the order of 0.1 cm\(^{-3}\), and values as high as 10 cm\(^{-3}\) have been reported, especially in young cirrus (Mace et al. 2001; Kärcher and Ström 2003). Even optically thin cirrus clouds may have ice particle concentrations of around 0.1 cm\(^{-3}\) (Mace et al. 2001).

Jensen and Pfister (2004) investigate the implications of small-scale variations in vertical wind velocity (and the associated variations in temperature) for the dehydration of air in the tropical tropopause layer. Interestingly, the properties of cirrus clouds are only weakly dependent on the synoptic-scale vertical wind (Mace et al. 2001), suggesting that vertical wind velocity on the small scale may play the significant role in defining these properties (Quante and Starr 2002).

In this study, we compare model results from the simulation of the nucleation and initial growth of ice particles with microphysical measurements made by
several instruments on board a DC-8 aircraft during the Subsonic Aircraft: Contrail and Cloud Effects Special Study (SUCCESS) campaign. The model is run along “trajectories,” that is, temperature–pressure time series. Temperatures have been calculated from vertical wind measurements made during SUCCESS and superimposed on a trajectory with a constant cooling rate typical of synoptic scales. These simulations show that in order to reproduce the observed number densities it is necessary to superimpose small-scale fluctuations on the synoptic trajectory. These small-scale temperature fluctuations are the result of gravity wave activity (Bae-meister et al. 1996) or turbulence (Quante and Starr 2002). In the next section, we briefly describe the measurement data used in this study, our model, and the simulations that were run. The modeled ice number densities and ice water contents are compared with corresponding measurements from SUCCESS in section 3, and section 4 contains a final discussion of the results and implications of the study.

2. Measurements and model setup

a. Types of trajectory simulations

Simulations of the microphysical properties of the measured cirrus clouds were carried out using a microphysical box model. This Lagrangian box model is designed to simulate the nucleation and growth process of ice particles. The details of the model are given in (Luo et al. 2003); however, a few central issues are discussed in this section.

The model simulates the conditions experienced by air parcels using temperature and pressure data along air parcel trajectories. Homogeneous freezing rates are calculated from the theory of Koop et al. (2000), assuming preexisting H₂SO₄/H₂O solution droplets. In our model, the sulphuric acid is regarded as nonvolatile. Because of the strong dependence of the nucleation rate on the H₂O concentration in the aerosols, we limit the change of H₂O concentration in the liquid phase during the numerical calculation by introducing a variable time step. We calculate the time step for each integration step ensuring that the maximum change of H₂O concentration in all size bins does not exceed a threshold value of 1%. Lowering this threshold value further does not lead to any change of the calculated ice number densities. The H₂O uptake coefficient on ice is assumed to be equal to 1 in the standard run and is tested for sensitivity later.

As an initial condition, the aerosol was assumed to be lognormally distributed with a mode radius of 0.025 μm, a half-width of σ = 1.8, and a number density of 800 cm⁻³ [a typical value measured by the cloud condensation nuclei (CCN) counter during the SUCCESS campaign]. The number density of aerosol particles is comparable with the value (1000 cm⁻³) used by Jensen and Pfister (2004).

The freezing probability of the aerosol particles is given by the homogeneous nucleation rate, which depends on the water activity of the droplets and temperature (Koop et al. 2000). As long as the aerosol droplets are not yet frozen, they will take up water and dilute during the cooling. The freezing probability is further proportional to the volume of the particles. One could also use this theory to model other droplet compositions with an aqueous phase such as H₂SO₄/NH₃/H₂O, and we would expect the results to be very similar (Koop et al. 2000). The volume of the aerosol depends on the water uptake by the aerosol particles, which may vary according to aerosol composition. However, the difference in the volume of aerosol particles due to a difference in water uptake is a second-order effect, and the resulting uncertainty in ice number density is much smaller than the uncertainty in the initial aerosol size distribution.

In the present study, we concentrate on the ice number density resulting from the nucleation event. After ice nucleation has occurred along a trajectory, we stopped the simulation once the ice saturation is reached due to H₂O condensation or temperature increase (i.e., we did not follow these air parcels to investigate the eventual evaporation of ice particles as a result of a temperature increase). The changes in ice number densities due to mixing, scavenging, and sedimentation were not taken into account. Therefore, caution should be used, when comparing our results with the subset of the measurements referring to large ice particles (r ≥ 20 μm). However, this affects only a small fraction of the measurements, and then mostly those of one of the four instruments, namely, the measurements made by the two-dimensional cloud probe (2D-C) (see section 3c).

Simulations were run using two different types of trajectories:

(i) Using the 40-yr European Centre for Medium-Range Weather Forecasts (ECMWF) Re-Analysis (ERA-40) data for 15 May. The temperature history of an air parcel was calculated, starting from a point on the flight path where cirrus measurements were performed. A total of 50 simulations were run, in which this single ECMWF trajectory was shifted up and down in temperature to provide a range of cooling rates at the nucleation temperature. Instead of modifying a single trajectory, we
have also run the model using a variety of different ECMWF-derived trajectories, but the final results are very similar.

(ii) Using trajectories with a small-scale temperature fluctuation, which describes the mesoscale activities not represented in large-scale models, superimposed on the ECMWF trajectory used in (i) or (for simplicity and without loss of accuracy) on a trajectory with a small constant cooling rate. We have done both in this work, but show, for better referencing, the results of the second approach using a constant cooling rate of 0.2 K h\(^{-1}\). The choice of this particular value has minimal effect on the results as the final cooling rate is dominated by the small-scale temperature fluctuation. A total of 4000 simulations were run for this type of trajectory, with randomly chosen phases of the temperature fluctuation overlaid on the trajectory with the constant cooling rate. The series of temperature fluctuations we used is not intended to represent particular temperature histories of specific air parcels, rather it is intended as a method of providing different cooling rates at the time of nucleation, at which point the number density of ice crystals in the resulting cloud is determined (see section 3). It should be noted that the series of temperature fluctuations superimposed on the underlying weakly varying trajectory may cause ice nucleation at more than one point on this trajectory (when a weakly nucleating cooling event is quickly followed by a stronger one). However, as a result of the drying of the gas phase following nucleation, in general, most of the subsequent fluctuations will come too late for additional nucleation. This leads to an important “shadowing effect,” which will be discussed further in section 3d.

The trajectories begin at a temperature of 229 K, pressure of 300 hPa, and water mixing ratio of 300 ppmv. These are mean values taken from the SUCCESS observations. As we will discuss in section 3a, the choice of initial temperature and pressure does not have a strong influence on the final ice number density.

b. Temperature fluctuations

The proper calculation of ice nucleation leading to cirrus cloud formation requires air parcel trajectories including the mesoscale temperature fluctuations, which are not explicitly represented in large-scale models (such as that of the ECMWF). For the modeling of the number density of ice particles, the cooling rate of the air parcel is the most critical parameter. It was shown by Bacmeister et al. (1996, 1999) that aircraft measurements of temperature or vertical winds may be used to analyze the gravity wave properties. However, the measured quantities cannot be used directly as air parcel trajectory information because

(i) the measurements are not taken along streamlines (i.e., in a quasi-Lagrangian sense), and
(ii) the streamlines of the air velocity field under the influence of gravity waves are in general not stationary (except for larger-scale fluctuations such as mountain waves).

As a result of these two effects, the wave activity in the natural environment is of lower frequency and amplitude than suggested by the measured time series of vertical winds or temperatures in the moving reference frame of the aircraft. Here, an analysis using Fourier transformations enables us to construct the time series of mesoscale temperature fluctuations in the inertial reference frame of the natural system (Bacmeister et al. 1996, 1999).

Observations of the vertical velocity \(w\) at various spatial positions \(x\) at time \(t_0\) can be used to derive the amplitude \(W\) in frequency space \(k\) by using the Fourier transform as follows:

\[
W(k) = e^{-i\omega_0 t_0} \int_{-\infty}^{\infty} w(x, t_0) e^{-i k x} dx. \tag{1}
\]

Here, \(x\) is the projection of the aircraft path onto the wind direction. The vertical wind can be obtained from the backward Fourier transform as follows:

\[
w(x, t_0) = \frac{1}{2\pi} e^{i\omega_0 t_0} \int_{-\infty}^{\infty} W(k) e^{i k x} dk.
\]

Assuming that the aircraft flies with a much higher speed than the group velocity of the intrinsic waves, which is usually the case, \(w(x, t_0)\) is simply the vertical wind measured on board the aircraft.

In a coordinate system \(x'\) moving with an air parcel with horizontal velocity \(u\), we have \(x = x' + ut_0\) and

\[
w(x', t_0) = \frac{1}{2\pi} \int_{-\infty}^{\infty} W(k) e^{i\omega_{t_0} + kx' + uit_0} dk.
\]

If \(x' = 0\) (we just follow one air parcel), and replacing \(t_0\) with \(t\), then

\[
w(0, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} W(k) e^{i\omega + uit} dk
\]

\[
= \frac{1}{2\pi u} \int_{-\infty}^{\infty} W(k) e^{i\omega + uk t} du.
\]
Replacing $\omega + uk$ by $uk'$ yields

$$\int_{-\infty}^{\infty} W(k' - \omega/u) e^{iuk't} du'$$

$$= F^{-1}\left[W(k' - \omega/u)\right]$$

$$= F^{-1}[G(k'u)].$$

(2)

Here $F^{-1}$ denotes the backward Fourier transform with a kernel $G$, which is equal to $W/u$.

In Eq. (2), when $w = 0$, we recover the case of stationary waves, that is, $G = W$. However, since the waves are not stationary we need to know the frequency distribution of waves representative for the troposphere during the SUCCESS flights that were not perturbed by orographic waves. We follow the suggestion made by Bacmeister et al. (1996) of equally distributed intrinsic frequencies between the Coriolis frequency $f$ and the Brun-Väisälä frequency $N$, which is equivalent to the $\omega^{2}$ dependency of the Fourier coefficients in the power spectral density of the temperature amplitude associated with the vertical wind fluctuations. The power spectrum $G^{2}$ integrated over this frequency range can then be calculated using

$$G^{2}(k'u) = \frac{1}{u^{2}} \int_{-\infty}^{\infty} \int_{\omega}^{\omega+N} W^{2}(k' - \omega/u) d\omega$$

(3)

for $uk' - \omega \geq 0$.

Again, here $u$ is the horizontal wind velocity. The power spectral densities (PSDs) $F_{f}$ of the temperature fluctuations can be obtained from $G^{2}$ (Bacmeister et al. 1999) as follows:

$$F_{f}(k'u) = T_{f}^{2} F(k'u) = \left(\frac{dT}{dz}\right)^{2} \frac{1}{(k'u)^{2}} G^{2}(k'u),$$

(4)

with the amplitude of the variance $T_{f}^{2}$ the normalized PSDs $F$, and the lapse rate $dT/dz$.

Using Eqs. (1), (3), and (4), the PSDs for vertical wind and temperatures can be calculated from the vertical winds measured by the aircraft. The reverse Fourier transform of the PSDs provides us with the vertical velocity and temperature time series for the air parcel [Eq. (2)]. We took all the measurements of vertical winds performed during SUCCESS for pressures <500 hPa to calculate the mean PSDs except those for 23 and 30 April and 2 and 4 May 1996, which aimed at studying wave cloud effects [see section 2c(2)]. The amplitude of the temperature fluctuations varies from flight to flight and also during the individual flights. The air parcel position [x in Eq. (1)] can be converted to air parcel time using the distance and the horizontal wind speed ($x/u$). Therefore, we performed the Fourier analysis over 6-h periods of air parcel time. The resulting mean temperature PSDs are shown in Fig. 1a, and the distribution of $T_{f}^{2}$ is shown in Fig. 1b. The mean temperature variance is 1.24 K$^{2}$.

The backward ECMWF trajectory used for the simulation for 15 May 1996 and associated cooling rates are shown in Fig. 2a. The temperature and cooling rates of air parcels with three different $T_{f}^{2}$ are shown in Figs. 2b–d. The distribution of the cooling rates from the temperature fluctuation is shown in Fig. 3. Here it can be seen that the temperature fluctuation mostly causes high cooling rates, the majority above 2 K h$^{-1}$, with some values higher than 30 K h$^{-1}$.

In the natural environment, slightly different air parcels, for example, with separations of ~100 m entering the cirrus cloud, will experience slightly different conditions due to inhomogeneities of the small-scale temperature field. To simulate this heterogeneity in our trajectory approach, a total of 4000 box model simulations were performed, using randomly chosen phases of the temperature variance in the backward Fourier transform and the weighting shown in Fig. 1b for the amplitudes.

**c. Measurements**

1) **INSTRUMENTATION**

The SUCCESS campaign took place in the United States, during April and May of 1996. Among other
objectives, it was intended to make measurements to better determine the microphysical and radiative properties of cirrus clouds and their formation mechanisms. A number of aircraft participated in the campaign. Here we use measurements made by instruments carried on the National Aeronautics and Space Administration (NASA) DC-8. The aircraft were based in Salina, Kansas, and flights were made over wide areas, including southern Kansas, northern Oklahoma, Colorado, and off the coast of Oregon. The instruments on the DC-8 carried out various microphysical measurements, including measurements of number density of ice particles (hereafter $n_{\text{ice}}$) and ice water content (IWC) that condensed in the cirrus clouds.

The instruments that were used to make the measurements are briefly described below:

(i) Multiangle Aerosol Spectrometer Probe (MASP) measures the particle size distribution optically. During the SUCCESS campaign, the detection range was from 0.15 to 12.5 $\mu$m in radius. Particles detected by MASP that were larger in radius than 12.5 $\mu$m were counted in the 12.5 $\mu$m bin (D. Baumgardner 2002, personal communication). In the analysis below, we excluded the particles in size bins smaller than 1 $\mu$m, as these channels might also include liquid particles.

(ii) Particulate Volume Monitor 100A (PVM-100A) is an optical probe that uses forward-scattered light from particles passing through a laser beam to measure IWC and particle surface area. From these values, the effective radius $R_e$ is also calculated (Gerber et al. 1994). The PVM can detect particles with radii up to a maximum of 25 $\mu$m (Young et al. 1998). From the PVM measurements, $n_{\text{ice}}$ can be estimated using the measured effective radius $R_e$ and the IWC.

(iii) Counterflow Virtual Impactor (CVI) measures IWC and number density of the residual solid particles after evaporating the ice particles that enter the instrument (Ström and Heintzenberg 1994). The counterflow prevents particles with an aerodynamic radius less than a lower cut size from entering the sampling nozzle (Twohy et al. 1997). For the flights that we consider here, the lower cut size is about 3–4 $\mu$m. The upper size of particles measured is only restricted by the size of the sampling nozzle (C. Twohy 2002, personal communication), but as discussed in section 3c, larger particles can introduce errors into the $n_{\text{ice}}$ measurements.

(iv) 2D-C is another optical probe that can detect particles with radii above ~25 $\mu$m (Heymsfield et al. 1998). The sample volume decreases with the square of the radius for radii below 80 $\mu$m, and the particle concentration for particles with radii below 50 $\mu$m is not very well determined (A. J. Heymsfield 2002, personal communication).

Figure 4 shows the sampling ranges used in the present study for the in situ particle instruments applied.
during SUCCESS. In addition to these four instruments, the DC-8 also carried particle imagers yielding qualitative information of shapes and sizes (not used in this study).

2) Data sampling

The microphysical measurements we use for comparison with our model results were made on the following days: 13, 15, 16, 18, 20, 21, 24, and 29 April and 7, 8, 10, 12, and 15 May 1996. We use all measurements made from the DC-8 except those indicated to have been taken in wave clouds (Toon and Miake-Lye 1998). We also exclude measurements with concentrations of NO$_y$ greater than 0.5 ppbv, as these air masses are likely to have been affected by deep convection (Fischer et al. 2002) or aircraft contrails (Campos et al. 1998). Furthermore, only those measurements where the CVI measured IWC $\geq$ 0.02 mg m$^{-3}$ have been included (in order to guarantee in-cloud sampling). A total of 300 min of measurements satisfy simultaneously these criteria. Thus, the focus of this study is on ice clouds forming in air masses that are subject to slow upwelling and not affected by deep convection, aircraft contrails, or lee waves.

3. Comparison of simulations and measurements

a. Dependence of ice number density on cooling rate

It is well known that the ice number density $n_{\text{ice}}$ established during a freezing event is highly dependent on the cooling rate at the time of the ice nucleation. In brief, high cooling rates sustain high supersaturations and lead to the nucleation of ice in many aerosol particles although the first ice particles have already started to deplete the gas phase. Conversely, this depletion under conditions of low cooling rates yields an early collapse of the supersaturation and hence the nucleation of only a few ice particles. However, systematic investigation of the effect of small-scale waves on the formation of cirrus clouds is still lacking. Kärcher and Ström (2003), based on aircraft-borne vertical wind measurements and observations of nuclei in evaporating ice particles obtained during cirrus cloud traversals in the interhemispheric differences in cirrus properties from anthropogenic emissions (INCA) campaign, show the importance of small-scale fluctuations for cirrus cloud formation. However, in their analysis they relied on applying the measured vertical wind velocities directly as instantaneous cooling rates, that is, they did not use a trajectory approach and hence ignored the effect of previous partial nucleation on subsequent nucleation (the shadowing effect discussed in section 3d). Furthermore, to obtain agreement with the observed data, they had to increase the upwelling within the clouds by up to 20 cm s$^{-1}$, which is a relatively strong change in the wind data (cf. the average additional ice resulting from such a procedure as indicated by Fig. 5 below). In the following, we apply trajectory analysis to the SUCCESS dataset and avoid any further manipulation. Very recently, Haag and Kärcher (2004) investigated the effects of heterogeneous ice nuclei on the INCA measurements and use high-frequency gravity wave signatures in the temperatures. In doing so they refer also to the analysis of the SUCCESS data in the present study.

For the simulations of $n_{\text{ice}}$ under the influence of temperature (and pressure) changes along air parcel trajectories, it is helpful to first investigate the dependence of the ice number density on a constant cooling rate for different freezing temperatures; see Fig. 5. In the case of rapid cooling (e.g., by $\sim$40 K h$^{-1}$), a relatively high final ice number density (6 cm$^{-3}$) is reached. In the case of slower cooling, the nucleation rate and hence the final ice number density, are lower (e.g., with a cooling rate of $\sim$3 K h$^{-1}$, $n_{\text{ice}}$ $\sim$0.1 cm$^{-3}$ is reached). In all these and the following calculations, the nucleation rate by Koop et al. (2000) for homogeneous ice
nucleation has been used. While Fig. 5 establishes the basic relationship between cooling rate and resulting ice number density, we emphasize that this relationship has only limited applicability in the real atmosphere.

b. Compensation between ice nucleation and growth

A major simplification of the present work derives from the fact that \( n_{\text{ice}} \) is strongly dependent on the cooling rate but reveals only a weak dependence on the absolute value of freezing temperature even under the quite diverse conditions typical for the natural atmosphere (see the curves in Fig. 5). This interesting behavior stems from a compensation between ice nucleation and growth: at higher altitudes in the troposphere, there is a lower sensitivity of the nucleation rate to a temperature change due to the lower ambient temperature (see Fig. 2b in Koop et al. 2000). At higher altitudes, the diffusion of the water molecules to the growing ice embryos is also faster, leading to an earlier quenching of nucleation. Both these effects would lead to a lower ice number density at higher altitudes. However, the \( H_2O \) partial pressure minus the \( H_2O \) vapor pressure over ice is much smaller at lower temperatures, leading to slower growth of ice crystals and a later quenching of the nucleation. This compensation justifies our choice of a constant freezing temperature of 226 K in the simulations. Note that this minimal dependence of \( n_{\text{ice}} \) on the actual freezing temperature is not in contradiction with the findings of Jensen and Toon (1994) and Kärcher and Lohmann (2002). They found considerable dependence of \( n_{\text{ice}} \) on the absolute temperature, but performed their simulations with different freezing temperatures at the same pressure level, not accounting for the atmospheric pressure/temperature relationship.

c. Comparison of observed and modeled ice number densities

The microphysical properties of ice particles in cirrus clouds were measured using several different instruments carried on board the DC-8 during the SUCCESS campaign.

In Fig. 6 we show the observations and the model results plotted as a histogram of frequency versus number density (\( n_{\text{ice}} \)). The model results have been filtered according to the detection ranges specified in Fig. 4; that is, the model results are shown as they would appear, had the modeled particle distributions been sampled by these instruments. In Fig. 6, \( df/dn_{\text{ice}} \) represents the fraction of measurements in a given number density bin.

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**Fig. 5.** Ice number density resulting from cooling at a prescribed constant rate (abscissa) corresponding to different water partial pressures and hence different freezing temperatures (line style). The choice of \((T, p)\) pairs leading to the different curves takes the natural occurrences of temperatures at certain levels in the troposphere into account. Under other conditions, for example, 300 hPa and 220 K, which are not of atmospheric relevance, deviations may occur (see dotted line).

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**Fig. 6.** (a), (e), (c), (g) A comparison between the \( n_{\text{ice}} \) measured by the four instruments MASP, CVI, PVM, and the 2D-C probe, and (b), (f), (d), (h) the simulated number densities from model runs using an ECMWF trajectory (dashed histogram) and a trajectory including gravity wave–induced temperature fluctuations (solid line histogram). Model results are filtered according to the detection characteristics of each instrument. For the CVI, measurements with IWC > 0.2 mg m\(^{-3}\) are included. For the PVM and the 2D-C, the sampling size range was simulated using the IWC data from the CVI (Fig. 8, histogram) as discussed in section 3e.
Although the $n_{\text{ice}}$ resulting from running the box model along the ECMWF temperature trajectory is within the range of the measurements of the 2D-C probe (Fig. 6g and dotted line in Fig. 6h), the correspondence with the $n_{\text{ice}}$ measured by MASP, PVM, and CVI is poor. The highest simulated $n_{\text{ice}}$ in the observational windows of these instruments along the ECMWF trajectories was approximately $0.02 \text{ cm}^{-3}$, which provides little overlap with the MASP measurements and no overlap with the PVM and the CVI measurements, which were higher than $0.05 \text{ cm}^{-3}$.

Far better agreement is seen between $n_{\text{ice}}$ from the fluctuating temperature simulations and the MASP measurements (see Figs. 6a,b), although the highest measured number densities can still not be fully explained (see discussion in section 3d and conclusions). Evidently, the high cooling rates given by the small-scale temperature fluctuations provide a straightforward explanation for the high number densities observed by the MASP. There is also substantially improved agreement between measured and simulated $n_{\text{ice}}$ for the PVM when using the fluctuating temperature trajectory. Furthermore, the correspondence between the simulation and the CVI measurements is greatly improved compared to the simulation run with a plain ECMWF trajectory. Possibly, the highest number densities measured by the CVI, as compared to the MASP or PVM, are related to the measurement principle of the CVI and could be explained by more than one residual being contained in a single ice particle, or by large particles colliding with the walls of the instrument and splitting (C. Twophy 2002, personal communication). The simulations of all instruments show a tail of distribution with low number densities ($<10^{-2} \text{ cm}^{-3}$), which are detected by the 2D-C probe but not by any of the other instruments (which have a cutoff at higher number density). This fraction of ice particles may grow to large sizes and sediment, thus contributing to the dehydration of the upper troposphere.

**d. Sensitivities of the model results**

To assess the effect of model parameters on our results, a sensitivity test was performed with the same aerosol distribution but a mode radius of 0.1 $\mu m$ (a factor of 64 more volume of aerosol particles). At small cooling rates (e.g., 1 K h$^{-1}$), the higher volume leads to a slight increase in ice number density ($\sim 6\%$, $n_{\text{ice}} \sim 0.02 \text{ cm}^{-3}$). At higher cooling rates (e.g., 40 K h$^{-1}$), the higher volume leads to a larger increase in ice number density (a 60% increase from $n_{\text{ice}} = 3.5$ to $5.5 \text{ cm}^{-3}$ at 40 K h$^{-1}$). Keeping the size and $\sigma$ constant and varying the total aerosol number density, a weak dependence of the final ice number density is apparent (an increase of 1% for 1000 $\text{cm}^{-3}$ aerosol particles and a decrease of 15% for 400 $\text{cm}^{-3}$ aerosol particles). Thus, the conclusions of the present simulation do not depend strongly on the assumption of the initial aerosol distribution.

Of higher importance than these sensitivities on quantitative changes are the qualitatively important sensitivities on the shadowing effect mentioned above and on the $\text{H}_2\text{O}$ mass accommodation coefficient, both of which we investigate next.

The dashed line in Fig. 7 shows the ice number density as calculated directly from the relationship between cooling rate and number density in Fig. 5, that is, without shadowing of smaller cooling rates by preceding nucleation events. This approach is similar to that used by Kärcher and Lohmann (2002); however, we note that the number density calculated by their method is a factor of 2 lower than when the comprehensive microphysical box model is used.

The difference in results between the two approaches may be explained by the fact that in reality, not all the cooling events along a trajectory have the chance to nucleate ice particles. The cooling events beyond a temperature minimum will be “shadowed” by a previous cooling event with lower temperatures. This effect can

![Fig. 7. Comparison of measured ice number density with sensitivity tests.](image-url)
be seen in Fig. 2d. A minimum temperature is reached at $t \approx 0.2$ h, and the following temperatures are always higher than this minimum. If nucleation occurs at this minimum, and the supersaturation is significantly reduced by uptake on ice particles, all subsequent cooling rates are irrelevant, while the simplified approach of Kärcher and Lohmann still takes them into account. It lies in the nonlinear nature of nucleation in small-scale turbulence that the locally lowest temperatures are often reached in events with the highest cooling rates, explaining the disproportionate importance of higher rates.

Finally, we note that there is a sensitivity of the ice number density with respect to the assumed H$_2$O mass accommodation coefficient on ice (Lin et al. 2002): decreasing the H$_2$O accommodation coefficient from 1 to 0.1 increases the ice number density by almost a factor of 5. The right-most dotted curve in Fig. 7 shows the simulated ice number densities for this case. Clearly, this does change the general conclusions of this study. Rather, there is now even better agreement with the measurements (shown for the case of MASP in Fig. 7). It is likely that under such cold conditions, the uptake coefficient of H$_2$O on ice is substantially smaller than unity, because the initialization of each new crystal plane requires a nucleation barrier to be surmounted. Of course, this is not a proof for reduced mass accommodation but reveals that such effects might indeed play a role.

e. Comparison of measured and modeled ice water content

Information on the IWC is provided by the 2D-C probe, the PVM, and the CVI, carried on board the DC-8. The IWC of those particles detected by the MASP has been estimated from the MASP data, assuming the particles to be spherical. Again, as these instruments have different detection ranges depending on particle size, the IWC provided by each instrument refers only to those particles in the corresponding radius range (see Fig. 4). The simulated IWC is shown in Fig. 8, where solid histograms show the simulation results when temperature fluctuations are used (except for the CVI) and dashed histograms show the simulation results when just the ECMWF trajectory is used. The solid lines show the IWC from the measurements made by each instrument.

The CVI measured the IWC when the ice particles were larger than 3–4 μm in radius. Most of the particles are larger than this cutoff size, even for the highest number densities because of the high total water (~300 ppmv). The measured IWC and number density from the other instruments depend on the size of the ice particles (see Fig. 4), which we have calculated from the respective simulated number densities and the IWC measurements made by the CVI (Fig. 8, CVI panel).

The modeled IWC needs to be normalized, as it depends upon the temperature relative to the frost point at the moment of the measurements. The H$_2$O partial pressure may differ from the vapor pressure over ice in a rapid cooling/warming event. Neither is known to us a priori. The measurements of IWC made by the CVI are suitable for this normalization as particles smaller than the 3–4-μm lower cutoff size contribute far less to the total IWC than larger particles; therefore, the IWC measurements from the CVI should be close to the actual IWC at the time of the measurement. The IWC measurements made by the CVI were smoothed (solid histogram in the CVI panel) and used, together with the simulated number density, to calculate the size of the simulated particles. This was done as follows: assuming spherical particles, the total IWC from each of the CVI measurements was divided among the total number of particles simulated by our model in a particular model run (assuming a monodisperse size distribution). When this is done for all CVI IWC measurements, it results in a distribution of particle sizes. The simulated IWC for the MASP, the PVM, and the 2D-C probe for that model run was then determined by sampling the par-
particles in this distribution according to the detection limits of those instruments. The process was repeated for all the bins in the number density distribution to give the IWC distributions shown in Fig. 8.

In all cases, the range of IWC calculated from the results of simulations using the trajectory with small-scale temperature fluctuations corresponds well with the range of measured IWC.

The total IWC simulated by the model is the same, regardless of which of the two trajectories are used; however, when the ECMWF trajectories are used, fewer particles are nucleated and the final particle size is much larger than when the fluctuating temperature trajectories are used. If some of the particles are large enough that they exceed the maximum size detected by a particular instrument, the IWC measured by that instrument will be correspondingly lower. For the CVI instrument, the IWC from the ECMWF trajectories and from the trajectories with temperature fluctuation are nearly identical, as it was assumed that all ice particles larger than 3–4 μm are seen by the CVI. The simulated IWC ranges from about 0.1 to 300 mg m$^{-3}$ once the measurements are smoothed using a running mean.

The simulated IWC matches the peak at high measured IWC reasonably well for the MASP, when temperature fluctuations are taken into account. In contrast to the reasonable agreement of the fluctuating temperature simulations, those runs using the unaltered ECMWF temperatures produced an IWC that was mostly too low (dashed curve).

In the case of the PVM instrument, whose detection efficiency was assumed to decrease for particles with radius >22 μm, most of the particles generated in the ECMWF temperature simulations are too large to be detected, resulting in a very low IWC, providing little correspondence with the measured IWC. The simulation with the fluctuating temperature trajectory, however, produced IWCs in good agreement with the measurements.

Finally, for the 2D-C probe, both sets of simulated results agree reasonably well with the measurements. The largest IWCs seen by the 2D-C probe remain unexplained but seem to contradict the CVI measurements.

4. Discussion and conclusions

Simulations of homogeneous ice nucleation were carried out using a comprehensive microphysical box model applied to trajectories obtained from ECMWF temperature and wind fields and to the same trajectories with small-scale temperature fluctuations superimposed. There have been a number of previous studies highlighting the potential importance of small-scale temperature fluctuations for cirrus cloud formation; however, these studies were either restricted to using a simple linear $n_{\text{ice}}(dT/dt)$ relationship (Kärcher and Ström 2003) or assumed fluctuations (Haag and Kärcher 2004) or were mainly model oriented (Jensen and Pfister 2004). In the present study, the temperature fluctuations have been derived from vertical wind measurements. Much higher number densities of ice crystals are obtained when small-scale temperature fluctuations are taken into account, as the number of nucleation events per volume of air is proportional to the cooling rate of the air parcel at the moment of the onset of ice nucleation. The atmospheric distribution of cooling rates is captured by running the simulation with 4000 different sets of small-scale temperature fluctuations superimposed on to the synoptic-scale trajectory. This produces a distribution of ice crystal number densities, which we have compared to cirrus cloud measurements performed during the SUCCESS field campaign. The ice crystal number densities from the simulations that account for the small-scale temperature fluctuations correspond reasonably well with the measurements, while the number densities simulated using the unaltered ECMWF trajectories are clearly too low.

The large range of cooling rates provided by the temperature fluctuations also helps to explain the variation of number densities over several orders of magnitude, which is apparent in cirrus observational data.

There is some discussion as to the reliability of the highest $n_{\text{ice}}$ measurements measured by various aircraft-based instruments; therefore we cannot rule out the possibility that some of the measurements shown in Fig. 6 are overestimated (Gayet et al. 1996, 2002). As can be seen in the CVI, PVM, and MASP panels of Fig. 6, a reduction in the measured number densities would indeed improve the match between our model results and the measurements. More reliable measurements would be extremely useful in future efforts to improve the understanding of formation mechanisms for ice clouds and their impact on climate.

The model does not take into account the possibility of some heterogeneous ice nucleation taking place before the onset of homogeneous nucleation. However, given the high cooling rates, which dominate the trajectories in this study (see Fig. 3), heterogeneous nuclei may not greatly reduce our modeled number densities. Furthermore, heterogeneous nucleation could certainly not have contributed significantly to the high ice number densities that were observed, for the following two reasons: First, insoluble particles that may induce heterogeneous nucleation have been measured on flights.
during SUCCESS on 21 April and 3 May 1996, with concentrations of about $2 \times 10^{-3}$ cm$^{-3}$ and $1 \times 10^{-2}$ cm$^{-3}$ at ice saturation, respectively (Rogers et al. 1998). Clearly, this is too low to explain the number densities within the cirrus clouds during SUCCESS (Fig. 6). Second, the presence of heterogeneous nucleation will not lead to the observed result of high number densities in the cirrus cloud. Rather, heterogeneous nucleation would always lead to a lower final number density of ice particles as compared to having only homogeneous nucleation, since the few heterogeneously nucleating particles absorb disproportionately much water and leave less for the subsequent homogeneous nucleation (DeMott et al. 1998). This point is in accordance with findings by Kärcher and Lohmann (2003) and Haag and Kärcher (2004), who termed this the negative Twomey effect.

In conclusion, we find that the only way to reach the measured number densities in the cirrus clouds observed during SUCCESS is to take the small-scale temperature fluctuations into account and to assume that the freezing of ice occurs homogeneously. Given the ubiquitous character of prerequisites leading to this result, it is of much more extensive applicability than just to the one campaign considered in detail here, as measurements of ice number densities in cirrus clouds reveal $n_{\text{ice}} = 0.1$ cm$^{-3}$ to occur commonly, with cases of $n_{\text{ice}}$ up to 10 cm$^{-3}$ also being reported. This requires homogeneous freezing subject to cooling rates of up to several 10 K h$^{-1}$, resulting from small-scale temperature fluctuations. These fluctuations remain unresolved even in high-resolution global models such as those of the ECMWF corresponding to T511 (or roughly $0.5^\circ \times 0.5^\circ$; Buizza et al. 2001). Other global circulation models and chemical transport models as used in climate and atmospheric chemistry studies have typical resolutions ranging from $2.8^\circ \times 2.8^\circ$ to $4^\circ$ (latitude) $\times 5^\circ$ (longitude; Chipperfield et al. 1996; Gupta et al. 2001; Rozanov et al. 1999) and are even less capable of describing the small-scale processes considered here. Such models will require physically justified cirrus cloud parameterizations if cloud development and precipitation and cloud radiative properties in the upper troposphere are to be described correctly. These parameterizations must take small-scale temperature fluctuations into account and must allow for the shadowing effect of temperature minima, if $n_{\text{ice}}$ is to be calculated directly from a distribution of cooling rates.

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