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Ice crystal concentrations in wave clouds: dependencies on temperature, $D > 0.5\,\mu m$ aerosol particle concentration and duration of cloud processing

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Abstract

Model equations used to either diagnose or prognose the concentration of heterogeneously nucleated ice crystals depend on combinations of cloud temperature, aerosol properties, and elapsed time of supersaturated-vapor or supercooled-liquid conditions. The validity of these equations is questioned. For example, there is concern that practical limitations on aerosol particle time-of-exposure to supercooled-liquid conditions, within ice nucleus counters, can bias model equations that have been constrained by ice nuclei (IN) measurements. In response to this concern, this work analyzes airborne measurements of crystals made within the downwind glaciated portions of middle-tropospheric wave clouds. A streamline model is used to connect a measurement of aerosol concentration, made upwind of a cloud, to a downwind ice crystal (IC) concentration. Four parameters were derived for 80 streamlines: (1) minimum cloud temperature along the streamline, (2) aerosol particle concentration (diameter, $D > 0.5\,\mu m$) measured within ascending air, upwind of the cloud, (3) IC concentration measured in descending air downwind, and (4) the duration of water-saturated conditions along the streamline. The latter are between 38 to 507 s and the minimum temperatures are between −34 to −14 °C. Values of minimum temperature, $D > 0.5\,\mu m$ aerosol concentration and IC concentration were fitted using the equation developed for IN by DeMott et al. (2010; D10). Overall, there is reasonable agreement among measured IC concentrations, IN concentrations derived using D10's fit equation, and IC concentrations derived by fitting the wave cloud measurements with the equation developed by D10.

1 Introduction

Ice nucleation is a pivotal process in the evolution of many cloud types (Braham and Squires, 1974; Cantrell and Heymsfield, 2005; DeMott et al., 2010; Murray et al., 2012). Ice crystals form via different pathways; the two fundamental distinctions are homogeneous and heterogeneous nucleation. Temperatures colder than −35 °C, and the

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existence of either haze particles or cloud droplets, are necessary conditions for the occurrence of the homogeneous pathway (Heymsfield and Miloshevich, 1993). Heterogeneous ice nucleation takes place on aerosol particles (ice nuclei, IN) and the known pathways are deposition, condensation freezing, immersion freezing and contact freezing (Vali, 1985; Murray et al., 2012). This work is focused on the latter processes.

Two contrasting approaches are used to translate measurements into equations used to predict IN activation, and thus ice crystal (IC) concentration, in cloud models. The first of these is diagnostic in the sense that IC concentration is formulated solely in terms of thermodynamic and aerosol state properties. The second is state and time dependent. In model intercomparison studies (Eidhammer et al., 2009; Niemand et al., 2012), these two frameworks produce significantly different IC concentrations. There are many reasons for these inconsistencies; fundamentally, they result because the time scale characterizing the development of a subcritical ice embryo into an ice crystal (Bigg, 1953; Vali and Stansbury, 1966), and how properties of an aerosol particle influences embryo development, are inadequately understood (Murray et al., 2012; Vali, 2014). Another relevant factor, but one which attenuates the framework-to-framework differences (Eidhammer et al., 2009), is that the Bergeron–Findeisen process can act to slow, or even shut down, the freezing nucleation pathways (i.e., condensation, immersion and contact freezing).

Our primary focus is the temperature- and aerosol-dependent IN fit equation developed by DeMott et al. (2010; hereafter D10). The D10 equation, hereafter Eq. (1), was developed with measurements of activated IN concentration derived using the continuous flow diffusion chamber (CFDC; Rogers et al., 2001). The IN measurements were made concurrently with measurements of the concentration of aerosol particles with diameter (D) larger than 0.5 µm ($n_{0.5}$)

$$N_{\mathrm{IN}}(T, n_{0.5}) = a \cdot (T_{\mathrm{o}} - T)^{b} \cdot (n_{0.5})^{c\cdot(T_{\mathrm{o}}-T)+d} \quad (1)$$

Here T is the temperature in the section of the CFDC operated above water saturation, T_{o} is a reference temperature (273.16 K), and a, b, c and d are the fitted coef-

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ficients. We reexamine Eq. (1) because it was developed with the CFDC operating in a manner which restricted the upper-limit diameter of aerosol particles processed within the CFDC ($D < 1.6\,\mu m$) and which restricted the duration of the particle's exposure to water-saturated conditions ($t < 10\,s$). Since both of these restrictions can cause the IN concentration to be underestimated (D10; Wright et al., 2013; DeMott et al., 2014), we use measurements made in and near clouds to evaluate the potential bias.

We have three specific objectives. First we use our airborne measurements of IC concentration to derive a temperature-dependent fit of those measurements. We refer to these two properties as N_{IC} and $N_{IC}(T)$, respectively. Specifically, we analyze IC concentration measurements recorded within the downwind (descending flow) portion of middle-tropospheric wave clouds, where IC concentration is thought to reflect IN activation that occurred upwind, within the colder and liquid-water saturated portion of the cloud. Second, we use our measurements to derive a temperature- and aerosol-dependent fit of N_{IC} based in Eq. (1). We refer to the latter as $N_{IC}(T, n_{0.5})$. Third, we analyze our measurements of N_{IC} with estimates of the interval of time an air parcel was exposed to water-saturation within a wave cloud. This is relevant to cloud modeling because many models employ a state- and time-dependent framework to predict IC concentration (e.g., Hoose et al., 2010). The IN, aerosol and IC concentrations relevant to our work are summarized in Table 1.

The foundations of our investigation are the wave cloud studies of Cooper and Vali (1981), Cotton and Field (2002), Eidhammer et al. (2010) and Field et al. (2012). Relevant to our work, these prior studies demonstrated that heterogeneous ice generation can be distinguished from both homogeneous nucleation, and from secondary ice production processes, and that the contribution from the deposition pathway is generally small in comparison to crystal production via the previously-mentioned freezing nucleation pathways.

Our investigation is most similar to the airborne studies of Eidhammer et al. (2010) and Field et al. (2012). Those authors analyzed cold-season (late fall) measurements

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made near, and within, middle-tropospheric wave clouds during the ICE-L project conducted in 2007. Their measurements were made over northern Colorado and southern Wyoming. Similar to that work, we sampled wave clouds along horizontal flight legs and also made no attempt to use the aircraft to follow the wave streamlines. Our airborne measurements were made in 2008 and 2009, in the same region as the ICE-L study, and also during the cold season (late winter and early spring). We analyze measurements made at locations where a streamline model indicated our aircraft intersected air that ascended into, and descended from, wave clouds. As we will discuss in detail, we develop a data set from eight flights; 80 wave cloud streamlines are analyzed. In contrast, Eidhammer et al. (2010) analyzed data from one flight, and modeled three streamlines. Field et al. (2012) expanded that analysis, and reported on measurement/model comparisons for 28 streamlines. In their analyses, both Eidhammer et al. (2010) and Field et al. (2012) exercised a streamline-following aerosol and cloud microphysical parcel model, and both derived the model's initial thermal state using measurements made downwind of the investigated wave clouds. In contrast, we use a streamline model to track the thermodynamic and time-distance evolution of air parcels (parcel microphysics is not evaluated), and we use measurements made immediately upwind of the investigated clouds, within ascending air, to initialize the model.

2 Measurements

We analyze airborne remote sensing and airborne in-situ measurements from the Wyoming Airborne Integrated Cloud Observation (WAICO) study conducted 2008 and 2009 (Wang et al., 2012). All measurements were acquired onboard the University of Wyoming King Air. The base of operations was Laramie, Wyoming. All of the sampled clouds were in the altitude range 3700 to 7400 m, and were located north of Laramie, within 110 km.

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2.1 Temperature and humidity

Temperature (T) was measured using a reverse-flow immersion thermometer (Lawson and Cooper, 1990). Dew point temperature (T_{dp}) was derived from vapor density measurements made with a LI-COR gas analyzer (model LI6262). The latter is characterized by a 0.2 s time response (Dobosy et al., 1997) and this value is somewhat smaller than the time response of the reverse-flow temperature sensor (~ 1 s; Rodi and Spyers-Duran, 1972). The inlet to the LI-COR was forward-facing and was operated subisokinetically with its inlet airspeed set at approximately 18 m s^{-1}. The latter is a factor of six smaller than the airspeed of the King Air (110 m s^{-1}).

2.2 Microphysics

Three wing-mounted optical particle counters are used in this analysis: (1) the Passive Cavity Aerosol Spectrometer Probe (PCASP), (2) the Forward Scattering Spectrometer Probe (FSSP), and (3) the Two Dimensional Optical Array Probe (2DC). Each of these was fabricated by Particle Measuring Systems (PMS; Boulder, CO).

The PCASP was used to measure the concentration of particles with diameters between 0.12 to 3.2 µm. Particle sizing was based on laboratory calibrations conducted using monodisperse test particles with refractive index $n = 1.59$ (Cai et al., 2013). PCASP concentrations were derived as the ratio of particle count rate divided by a calibrated sample flow rate (Cai et al., 2013).

Adiabatic compression warms the aerosol stream as it approaches the PCASP inlet. Strapp et al. (1992) estimated that this process occurs over 0.2 s. Once the stream reaches the probe, it is warmed by three anti-ice heaters (Snider et al., 2014). The time scale for diabatic (anti-ice) heating is approximately an order of magnitude smaller than the 0.2 s adiabatic warming. Because of both the adiabatic and diabatic processes, unactivated cloud droplets (haze particles), and cloud droplets, are partially evaporated prior to sizing within the PCASP. In the case of haze particles, evaporation is complete

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if the initial particle diameter is smaller than ∼1 µm (Strapp et al., 1992; Snider and Petters, 2008).

The FSSP was used to categorize cloud droplets sizes from 1.5 to 47.5 µm into 15 bins. During WAICO the cloud droplet concentrations were less than 300 cm^{-3}, so the FSSP dead time and coincidence errors are less than 25 % (Baumgardner et al., 1985). Both of these effects were accounted for in the data processing. Because our FSSP measurements come from clouds containing ice, bias due to ice crystal shatter also needs to be addressed. Since we only analyze FSSP measurements recorded near the upwind edge of the clouds, where the ice crystals are small (< 100 µm) and their concentration is low (< 0.4 L^{-1}), the effect of shatter on the FSSP measurements is not expected to be significant (Gardiner and Hallett, 1985; Gayet et al., 1996; Field et al., 2003) and was not evaluated.

Ice crystals were sized and counted using an optical array probe (2DC) (Pokharel and Vali, 2011). This instrument records a crystal as a two-dimensional image. Some images were rejected using criteria described in Pokharel and Vali (2011). Images which passed the rejection tests were sized in the along-track direction (hereafter, this dimension is termed "diameter") and these were binned into channels with lower-limit diameters set at 25, 50, 100, 150, 200, 250, 300, and 400 µm for the smallest eight of 20 channels; nearly all crystals recorded during WAICO classified into these eight channels. Because even the largest crystals in this set are smaller than the size known to shatter when impacted at aircraft velocities (Korolev and Isaac, 2005; Korolev et al., 2013), the effect of shatter was ignored. Concentrations were derived with the assumption that the optical depth of field was independent of crystal size and equal to the 2DC's sampling aperture (61 mm) (Vali et al., 1981).

2DC-derived concentrations were validated by Cooper and Saunders (1980). The basis for their validation was airborne 2DC concentrations measured simultaneous with concentrations derived by impacting ice crystals onto oil-coated slides (OCS) exposed in a decelerator. Crystals impacted on the slides were photographed and counted, the counts were increased by dividing by a size-dependent impaction efficiency, and

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diameter-integrated concentrations were computed for crystals with maximum dimension larger than 50 µm. The OCS concentrations were compared to 2DC concentrations. The latter were derived by integrating from 50 µm to larger diameters. Cooper and Saunders reported 2DC-OCS concentration ratios between 3.6 and 0.6 ($\overline{x} = 1.7$, $\sigma = 0.9$, number of samples = 12). From the comparisons it was concluded that, for crystals larger than 50 µm, the 2DC is capable of making quantitative concentration measurements.

Based on the findings discussed in the previous paragraph we derived N_{IC} (Table 1) as the diameter-integrated concentration corresponding to $D > 50$ µm. Further, we excluded from our analysis instances when the concentration of crystals in the first 2DC channel (25 to 50 µm) exceeded more than 50 % of the overall ($D > 25$ µm) diameter-integrated concentration. The intent of this criterion is avoidance of crystals whose concentration is uncertain because their depth of field is ambiguous. If we had summed those crystals into N_{IC}, the relative concentration bias could have approached a limiting value equal to the ratio of the 2DC manufacturer's recommendation for a 25 to 50 µm particle's depth of field (~ 4 mm) divided by the sampling aperture (61 mm) (Strapp et al., 2001).

For both the PCASP and the 2DC the relative Poisson sampling error was evaluated as the reciprocal of the square root of the particle count.

2.3 Air motion

Vertical and horizontal air velocities were derived from differential pressure measurements made at the tip of the King Air's nose boom (Parish and Leon, 2013).

2.4 Lidar

The upward-pointing Wyoming Cloud Lidar (Wang et al., 2009, 2012) was used to remotely sense cloud boundaries. The lidar transmits in the near ultraviolet (λ = 0.355 µm) at a pulse repetition frequency of 20 Hz. Seven lidar shots were averaged,

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making the effective sample rate ~3 Hz. The vertical resolution of the lidar is 3.75 m. Using the lidar measurement of attenuated backscatter and depolarization, we evaluated the boundaries between clear air and liquid cloud, and between liquid-dominated and ice-dominated cloud (Wang and Sassen, 2001).

In the next section we describe our determinations of the air parcel streamlines and how the lidar-derived cloud boundaries were used to evaluate the time interval, along the streamlines, within the liquid-dominated portions of the clouds.

3 Analysis

3.1 Parcel streamlines and parcel thermodynamic state

Here we explain how the streamlines were derived from measurements made during level-flight penetrations of 35 wave clouds. In our data set we have 19 penetrations made along the wind, and sixteen penetrations made against the wind. Also described is the parcel model we used to evaluate thermodynamic properties along the streamlines.

An average horizontal wind speed ($\overline{u}$) was derived from airborne in-situ wind measurements made during each of the cloud penetrations. That average was applied as a constant in our streamline analysis. In contrast, the in-situ measured vertical wind component (w) was oscillatory, so we fitted it as a sinusoid function, vs. along-track distance (x), and we assumed that the fitted vertical wind component ($w(x)$) did not vary vertically. Figure 1a shows the measured and fitted values of the vertical wind for a penetration that we showcase to illustrate our methods.

Within the ascending portion of the wave structure (e.g., to the left (upwind) of x = 10.5 km in Fig. 1a), we initialized several streamlines. The streamline center points were separated by ~ 550 m along the flight track (five seconds at 110 m s^{-1}). For each of the center points the 1 Hz measurements of T, T_{dp}, and pressure (P) were used to derive five-second averaged values of T, T_{dp}, and P. These three properties were used

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to fix an air parcel's initial thermodynamic state. A closed parcel model, conserving potential temperature below the lifted condensation level (LCL), and equivalent potential temperature, above the LCL, was used to evaluate the thermal state, along the streamline. Using this model, and the aforementioned descriptions of the horizontal and vertical wind components, we simulated the thermal and kinematic evolution of streamline-following air parcels. One of the evaluated relationships is the parcel's temperature as a function of the along-track distance. This is shown, for the example, in Fig. 1d. Also indicated are the minimum streamline temperature (T_{low}) and the measurement of temperature (red circle) made at the downwind intersection of the flight track and the streamline.

We compared our streamline temperatures, each evaluated at the downwind track-streamline intersections, and the corresponding measured temperatures. The average absolute difference is 0.3 °C (number of samples = 80). This agreement is consistent with a small effect, smaller than the temperature measurement error (±0.5 °C), coming from violations of either the closed parcel assumption or the assumptions of vertically-uniform $w(x)$ and constant $\overline{u}$.

3.2 Mixed-phase time

The interval of time an air parcel experiences water-saturated conditions was evaluated by combining the lidar measurements with the streamline information. We refer to this time interval as the mixed-phase time (t_{MP}). Figure 1b and c illustrate how t_{MP} was evaluated. At the upwind cloud edge, at $x = 9.5$ km but above the aircraft, the streamline encounters the first of two cloud boundaries. Using lidar measurements, we defined this upwind cloud boundary by its increased lidar backscatter and decreased lidar depolarization (compared to the depolarization in clear air). Approximately four km downwind, the streamline encounters the second boundary. We defined this boundary by its decreased lidar backscatter and increased depolarization. Here the boundary is between liquid- and ice-dominated cloud. Further, we defined t_{MP} as the integral of the parcel transit time between these two boundaries. For a few of the streamlines,

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the downwind track-streamline intersection was within the liquid-cloud region. In those cases, the calculation of t_{MP} was stopped at the intersection. The lower and upper bounds of t_{MP} are 38 to 507 s; the average t_{MP} is 221 s.

We obtained good agreement between values of t_{MP}, based exclusively on lidar, and those based partially on the in-situ measurements of T and T_{dp}. These comparisons were made by differencing the lidar-derived t_{MP} and a mixed-phase time derived using T- and T_{dp}-dependent determinations of the LCL (Sect. 3.1) combined with lidar-based determinations of the downwind cloud boundary. In this comparison the average absolute difference is 22 s. Each absolute difference was converted to a relative difference by dividing by the lidar-derived values of t_{MP}. The relative differences range from 0.0 to 0.9.

3.3 Aerosol particles and cloud droplets

In this section we evaluate the connection between upwind aerosol concentrations and in-cloud droplet concentrations. For each of the 35 cloud penetrations we evaluated five-second averages of the PCASP and FSSP concentrations. For the PCASP, the averaging interval was started five seconds upwind of the cloud, and for the FSSP, the averaging interval was started at the cloud edge. Averaging intervals are shown at the bottom of Fig. 2b and at the top of Fig. 2d. Also presented (Fig. 2a–c) are the size-resolved concentrations from the PCASP, FSSP and 2DC. The series shown in Fig. 2 are for the same section of flight illustrated in Fig. 1.

Similar to Eidhammer et al. (2010), we compared the upwind aerosol particle concentration ($D > 0.25$ µm; five-second averaged) to the in-cloud droplet concentration ($D > 1.5$ µm; five-second averaged). From the series presented in Fig. 2d, it can be seen that droplets, measured at ∼x = 11 km (i.e., downwind of the cloud edge), were more abundant than aerosol particles measured at $\sim x = 10.5$ km (i.e., upwind of the edge). Following this same averaging procedure, we evaluated a droplet-to-aerosol ratio for 32 of our 35 penetrations; three of the 35 were discarded because the droplets were smaller than the minimum size detectable by the FSSP ($D = 1.5$ µm). In the 32

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comparisons, the droplet-to-aerosol concentration ratios were consistently greater than 0.7. These results are consistent with the findings of Eidhammer et al. (2010). A reasonable inference is that the $D > 0.25\,\mu m$ particles are internally mixed, that the mixture's water-soluble fraction promoted the nucleation of the droplets, and that the mixture's water-insoluble fraction promoted ice nucleation, presumably via the condensation and immersion freezing pathways. The effect of ice nucleation on cloud properties is clearly evident at the downwind track-streamline intersection (at $\sim x = 15\,km$), in Fig. 1c (lidar depolarization), and in Fig. 2d (diameter-integrated crystal concentration).

3.4 $D > 0.5\,\mu m$ aerosol particle and IC concentrations

In addition to the $D > 0.25\,\mu m$ aerosol concentrations, analyzed in the previous section, we also evaluated $n_{0.5}$ (Sect. 1). These were averaged over the five-second time windows used for thermodynamic-property averaging (Sect. 3.1). For the rest of the paper, $n_{0.5}$ is reported as a particle count per standard cubic centimeter ($sccm^{-1}$). Also for the rest of the paper, values of N_{IC} (Table 1) are derived as five-second averages evaluated at the downwind track-streamline intersections (e.g., at $\sim x = 15\,km$ in Fig. 1c), and these are reported as a crystal count per standard liter (sL^{-1}).

3.5 Data set

In the previous sections we described how values of N_{IC}, $n_{0.5}$, T_{low}, and t_{MP} were evaluated for each streamline. The subset $\{N_{IC}, n_{0.5}, T_{low}\}$ is the streamline data we used to develop a fit of N_{IC}, according to the mathematical form of Eq. (1). However, before fitting our measurement data, we excluded streamlines affected by four effects: (1) an abundance of crystals in the first 2DC channel, (2) homogeneous freezing, (3) crystal sublimation, and (4) variable aerosol particle and crystal concentrations. Conditions for data *inclusion* are: (1) $N_{IC,D<50}/N_{IC,D>25} < 0.5$ (Sect. 2.2), (2) $T_{low} > -35\,^{\circ}C$ (Heymsfield and Miloshevich, 1993), (3) ice saturated, or larger relative humidity, at the downwind track-streamline intersection, and (4) relative Poisson sampling errors

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(Sect. 2.2) less than specified thresholds[1]. Out of the 116 streamlines we analyzed, 80 satisfy our data inclusion criteria. The set $\{N_{IC}, n_{0.5}, T_{low}, t_{MP}\}$ is provided for the 80 streamlines in the Supplement.

4 Fitted N equations

In this section we show results from fitting our measurement data with both temperature-dependent, and temperature-aerosol-dependent, equations. We start with a solely temperature-dependent fitting equation because many previous cloud modeling studies were based on such a relationship (e.g., Meyers et al., 1992), and because the rate of change of crystal concentration with temperature can have a profound impact on modeled cloud properties (Eidhammer et al., 2009).

We develop the fitting equations using logarithm-transformed crystal and logarithm-transformed aerosol concentrations. The reason for log-transforming the measurements is that we expect errors, in both crystal and aerosol concentration, to be multiplicative in the sense that larger values correspond with larger error and vice versa. Multiplicative error, scaling in proportion to the square root of concentration as predicted by the Poisson probability law (Young, 1962; Rogers and Yau, 1989), was documented by Cai et al. (2013) in their investigations of the PCASP's response to steadily-generated monodisperse test particles.

Figure 3a shows the temperature-dependent fit (i.e., $N_{IC}(T_{low})$) plotted vs. N_{IC}. The square of the Pearson correlation coefficient (r^2), for this scatter plot, is relatively small and demonstrates that temperature alone, via the fit equation, can only explain 44 % of the N_{IC} variability.

[1]The relative Poisson error thresholds adopted for IC concentration and for $n_{0.5}$, were 0.4 and 0.7, respectively. These values cut the distributions of the relative Poisson errors at their 99th percentiles.

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In Fig. 3b we plot our 80 fitted values of $N_{IC}(T_{low}, n_{0.5})$ vs. N_{IC}. Results shown here are for one of two fitting methods we implemented. In fit method #1 we used the Matlab Curve Fitting Toolbox (The MathWorks, Natick, MA), with the log-transformed version of Eq. (1), and derived the logarithm of a ($\ln a$), and the values of b, c and d. We also fitted our 80 measurements of $\{N_{IC}, n_{0.5}, T_{low}\}$ using the procedure described in D10. We refer to the latter as method #2. The advantage of method #1 is that it shortens D10's three-step procedure to one step. Another difference is that the number of points used to evaluate statistical error, associated with the fit coefficients, is relatively small in the case method #2. In method #1 the number of points is 80, while in our application of method #2 only four points were fitted in the second and third steps of D10's procedure.

The fit coefficients derived by D10, our fit coefficients (methods #1 and #2), and the method #1 and #2 statistical errors, expressed as standard deviations, are presented in Table 2. Focusing on results obtained using method #1, our coefficients $\ln a$ and b, and our coefficients c and d, are seen to agree within one and two standard deviations of D10's, respectively. Also, there is agreement, within one standard deviation, between our application of method #2 and D10's. It is also apparent that larger statistical error is evident for $\ln a$ and b derived in method #2, compared to method #1. This is because of the smaller number of points fitted in method #2, as discussed in the previous paragraph.

By inputting the statistical errors from Table 2 into a propagation of error equation (Young, 1962; their Eq. 13.9), we evaluated contributions to the relative variance of the logarithm of $N_{IC}(T_{low}, n_{0.5})$ (method #1). For $n_{0.5} \leq 3.4\,\mathrm{sccm}^{-1}$ (the average for our data set), and for temperatures over the full range of our data set ($-34 \leq T_{low} \leq -14\ ^{\circ}\mathrm{C}$), the relative variance is controlled by terms proportional to both the square of the statistical error in $\ln a$ and the square of the statistical error in b. Further, we also evaluated the fractional standard deviation of $N_{IC}(T_{low}, n_{0.5})$ (method #1). For the same $n_{0.5}$ and T_{low} settings provided above, the fractional standard deviation is ~ 4 and increases to ~ 5 if $n_{0.5}$ is set to $16\,\mathrm{sccm}^{-1}$ (the maximum for our data set). Yet, in spite of this uncertainty, our fitted (method #1) and measured values are seen to correlate over three decades

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of IC concentration (Fig. 3b). Also illustrated are fitted concentrations, derived using Eq. (1) with D10's coefficients and our measurements of T_{low} and $n_{0.5}$ In either case the r^2 is ∼ 0.7 and thus larger than that for the temperature-only fit (cf., Fig. 3a).

We also evaluated the fraction of the measured crystal concentrations that plot within a factor of two of the fit. Based on our method #1 coefficients, this percentage is 69 % and thus larger than the percentage (66 %) based on fit coefficients from D10 (the percentage is 60 % when using the method #2 coefficients; not shown here). Thus, we obtained better fitted-vs.-measured agreement with our method #1 fit coefficients, and poorer agreement with either our method #2 coefficients or with the D10 coefficients.

5 Effect of mixed-phase time

As was discussed in the introduction, there is an outstanding question in atmospheric science community regarding the time-dependent nature of ice nucleation. Of relevance for our data set, with its average $t_{MP} = 221$ s (Sect. 3.2), is the possibility that the characteristic time for a subcritical ice embryo to transition to a detectable ice particle is comparable to t_{MP}. If that were the case, we would expect that streamlines associated with larger mixed-phase times, all other things equal, would have larger IC concentrations. We explored this by stratifying our 80 determinations of $\{N_{IC}, n_{0.5}, T_{low}, t_{MP}\}$ into four T_{low} subsets. In Table 3 we present the subset's minimum and maximum temperatures, the averaged $n_{0.5}$, and the number of data values. For each of these we tested the hypothesis that $\ln(N_{IC})$ is correlated with $\ln(t_{MP})$. Values of the Pearson correlation coefficients (r), and the levels of significance (p), demonstrate that none of the correlations are significant (i.e., all have $p > 0.05$). This same conclusion was reached after removing from the correlations those points exhibiting the largest t_{MP} uncertainty (relative difference > 0.3, Sect. 3.2), but those results are not shown in Table 3. We also stratified by $n_{0.5}$ within the four T_{low} subsets. One of those correlations ($\ln(N_{IC})$ vs. $\ln(t_{MP})$) approaches statistical significance, with $p = 0.1$ and with 10 paired values; the

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rest have $p > 0.1$. That subset plots in the gray rectangle shown in Fig. 4a and the N_{IC} vs. t_{MP} correlation for that subset is shown in Fig. 4b.

In spite of these suggestions of a connection between crystal concentration and mixed-phase time we cannot argue convincingly that time-dependent effects were significant for crystals within the clouds we studied. Our ability to argue for, or against a dependence on t_{MP}, was limited by the number of points within the analyzed data subsets. Thus, in future wave cloud studies, attention should be paid to strategies which generate an adequate number of points within specified temperature and aerosol ranges.

6 Summary and conclusion

The result we present in Table 2, with fit coefficients generally consistent, in a statistical sense, with those reported by D10, is significant because it validates D10's equation using different methodology. In short, our technique uses a streamline model to connect a measurement of aerosol concentration ($n_{0.5}$), made upwind of a wave cloud, to a downwind measurement of IC concentration. Our reconfirmation of the connection between crystals and $n_{0.5}$ – the connection implied by Eq. (1) – is conceptually appealing because it acknowledges that aerosol particles are necessary for the occurrence of heterogeneous ice nucleation. Appeal also comes from the linkage provided by Eq. (1), through aerosol, to cloud processes.

We also probed the conjecture that the duration of nuclei exposure to water-saturated conditions is a determinant of IC concentration. Our analysis shows no statistically-robust evidence for this. This finding is relevant to descriptions of ice nucleation within water-saturated layer clouds (e.g., stratocumulus and altostratus) where temperature is relatively uniform, and steady, and where time-dependent ice nucleation is suspected of occurring continuously and with substantial meteorological impact (Crosier et al., 2011; Westbrook and Illingworth, 2013). In fact, many model representations of heterogeneous nucleation anticipate this time-dependent, constant-temperature, phenomenon.

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Also, in some models, the nucleation rate is set to zero when the temperature tendency is zero or positive (Khain et al., 2000; Muhlbauer and Lohmann, 2009), but this action is not supported by all of the experimental evidence currently available (for a review, see Vali, 2014). Further investigation is needed to confirm our conclusion of little, if any, time-dependent effect within the cloud type we studied (middle-tropospheric wave clouds). Going forward, we anticipate our methodology will help advance understanding of time-dependent atmospheric ice nucleation, and atmospheric ice nucleation in general.

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Table 1. Symbols used to represent aerosol, IN and IC concentrations.

Symbol	Definition	Dimension
$n_{0.5}$	Measured aerosol concentration ($D > 0.5\,\mu m$)	$sccm^{-1}$ [a]
N_{IC}	Measured IC concentration ($D > 50\,\mu m$) [b]	sL^{-1} [c]
$N_{IC}(T)$	Temperature-dependent fit of IC concentration (see Sect. 4)	sL^{-1}
$N_{IC}(T, n_{0.5})$	Temperature- and aerosol-dependent fit of IC concentration (see Sect. 4)	sL^{-1}
$N_{IN}(T, n_{0.5})$	Temperature- and aerosol-dependent fit of IN concentration (D10) (see Eq. 1)	sL^{-1}

[a] Aerosol particle count per standard cubic centimeter at $P = 1.013 \times 10^5$ Pa and $T = 273.15$ K;
[b] 2DC concentration for crystals sizing larger than 50 µm (see Sect. 2.2);
[c] Particle count per standard liter at $P = 1.013 \times 10^5$ Pa and $T = 273.15$ K.

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Table 2. Equation (1) fit coefficients.

Coefficients	Fit D10[a]	Fit Method #1	Statistical Error[b] Method #1	Fit Method #2	Statistical Error[c] Method #2
ln *a*	−9.73	−14.89	2.93	−8.67	6.65
b	3.33	4.79	0.89	2.86	2.21
c	0.0264	0.0076	0.0313	0.0225	0.027
d	0.0033	0.86	0.89	0.49	0.68

[a] Fit coefficients from D10;
[b] The standard deviations for coefficients fitted via method #1;
[c] The standard deviations for coefficients fitted via method #2.

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Table 3. T_{low} subsets and the $\ln(N_{IC})$ vs. $\ln(t_{MP})$ correlations.

T_{min}	T_{max}	$\overline{n_{0.5}}$	Number of samples	r[a]	p[b]
−34	−29	5.50	20	0.20	0.20
−29	−24	2.88	30	0.21	0.13
−24	−19	3.50	15	−0.05	0.57
−19	−14	2.57	15	0.06	0.44

[a] The Pearson correlation coefficient for the regression of $\ln(N_{IC})$ vs. $\ln(t_{MP})$;
[b] Level of significance, values of this parameter greater than $p = 0.05$ indicate an insignificant correlation.

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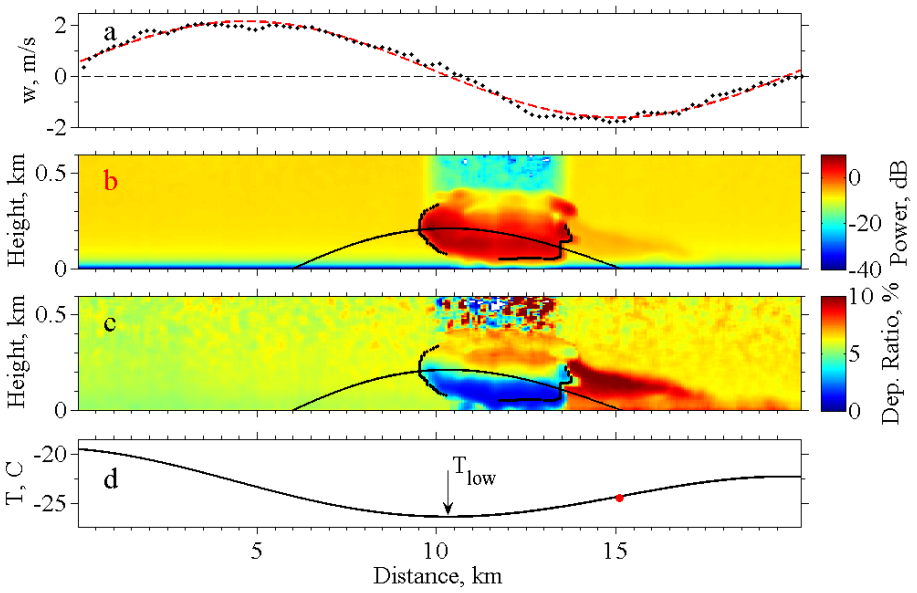

Figure 1. Airborne level-flight sampling a few tens of meter below a wave cloud between 18:17:45 and 18:20:09 on 27 February 2008. Airflow is from left to right. **(a)** In-situ vertical velocity measurements and the sinusoid fit. **(b)** The example streamline (black) overlain on lidar backscattered power; the two other black lines delineate the liquid-cloud and ice-cloud boundaries discussed in the text. **(c)** Example streamline overlain on lidar depolarization ratio; the two other black lines delineate the liquid-cloud and ice-cloud boundaries discussed in the text. **(d)** Streamline temperature, minimum streamline temperature, and the in-situ measured temperature at the downwind track-streamline intersection (red circle).

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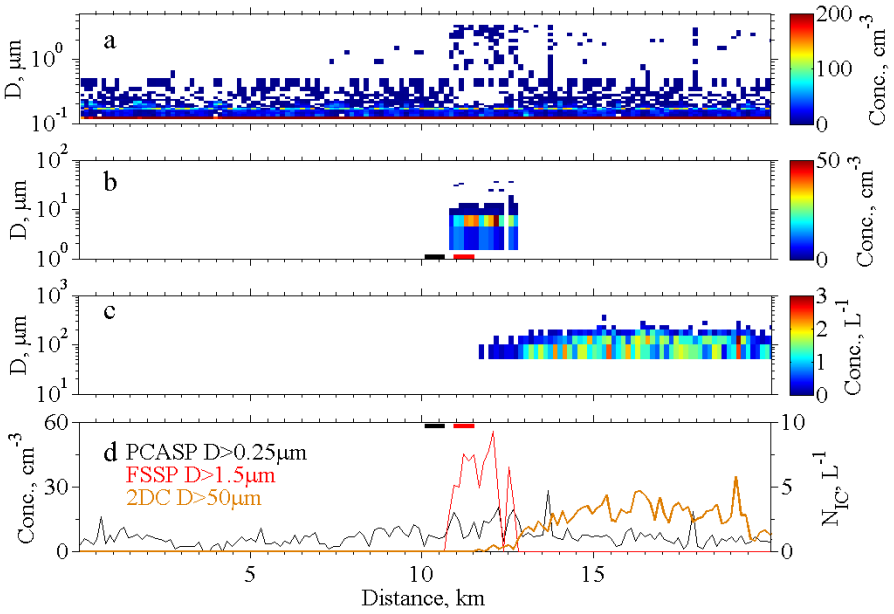

Figure 2. The same segment of flight as shown in Fig. 1; **(a)** Size-resolved PCASP concentrations. **(b)** Size-resolved FSSP concentrations. The black and red horizontal rectangles at the bottom of this panel are the five-second averaging intervals for aerosol and droplets analyzed in Sect. 3.3. **(c)** Size-resolved 2DC concentrations. **(d)** Diameter-integrated PCASP ($D > 0.25\,\mu m$, black line), diameter-integrated FSSP ($D > 1.5\,\mu m$, red line), and diameter-integrated 2DC ($D > 50\,\mu m$, orange line) concentrations. Averaging intervals for aerosol and droplets are repeated from panel b.

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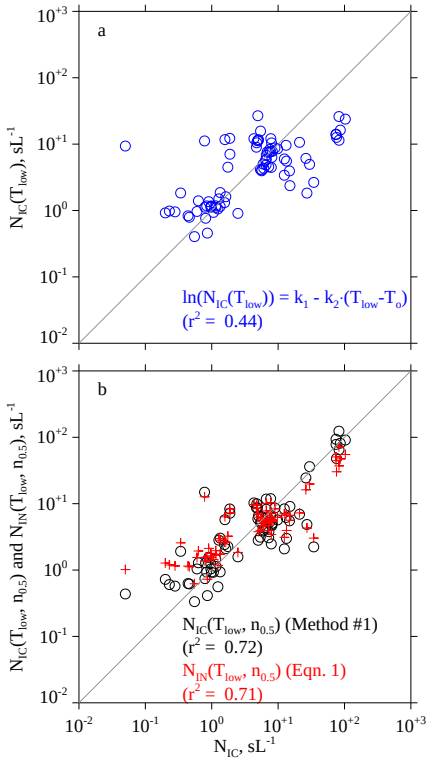

Figure 3. (a) Values of $N_{IC}(T_{low})$ ($\ln(N_{IC}(T_{low})) = k_1 - k_2 \cdot (T_{low} - T_o)$ with $k_1 = -3.93$ and $k_2 = 0.22\ ^\circ C^{-1}$) plotted vs. N_{IC}. **(b)** As in Fig. 3a, but with $N_{IC}(T_{low}, n_{0.5})$ (method #1 fit coefficients), and $N_{IN}(T_{low}, n_{0.5})$ (Eq. 1), plotted vs. N_{IC}. In Fig. 3a and b, the square of the Pearson correlation coefficients (r^2) was evaluated using log-transformed concentrations. Also, the one-to-one line is shown in both panels.

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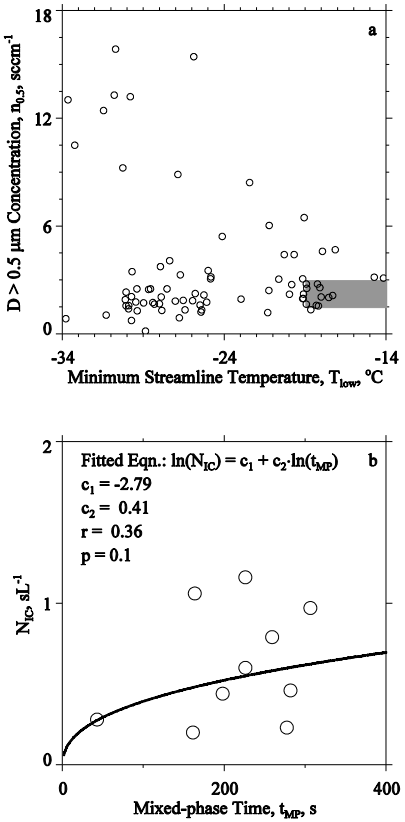

Figure 4. (a) The 80 paired values of $n_{0.5}$ and t_{MP} in our data set. The gray rectangle highlights the 10 points in the subset defined by $-19 \le T_{low} < -14$ °C and $1.5 \le n_{0.5} < 3.0\,\mathrm{sccm}^{-1}$. **(b)** The 10 paired values of N_{IC} and t_{MP} from the gray rectangle shown in Fig. 4a. The black line is the fitting equation $\ln(N_{IC}) = c_1 + c_2 \cdot \ln(t_{MP})$. The Pearson correlation coefficients (r), and the level of significance (p), were evaluated using the log-transformed concentrations and log-transformed mixed-phase times.

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