1 INTRODUCTION

The Cirrus Parcel Model Comparison (CPMC) is a project of the GEWEX Cloud System Study Working Group on Cirrus Cloud Systems (GCSS WG2). The primary goal of this project is to identify cirrus model sensitivities to the state of our knowledge of nucleation and microphysics. Furthermore, the common ground of the findings may provide guidelines for models with simpler cirrus microphysics modules.

Table 1: Simulation identifiers.

<table>
<thead>
<tr>
<th>$W$ [m/s]</th>
<th>0.04</th>
<th>0.2</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td>HN-ONLY</td>
<td>Ch004</td>
<td>Ch020</td>
<td>Ch100</td>
</tr>
<tr>
<td></td>
<td>Wh004</td>
<td>Wh020</td>
<td>Wh100</td>
</tr>
<tr>
<td>ALL-MODE</td>
<td>Ca004</td>
<td>Ca020</td>
<td>Ca100</td>
</tr>
<tr>
<td></td>
<td>Wa004</td>
<td>Wa020</td>
<td>Wa100</td>
</tr>
<tr>
<td>HN-$\lambda$-fixed</td>
<td>Ch020L</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Wh020L</td>
<td></td>
<td></td>
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</table>

We focus on the nucleation regimes of the warm (parcel starting at $-40^\circ$C and 340 hPa) and cold ($-60^\circ$C and 170 hPa) cases studied in the GCSS WG2 Idealized Cirrus Model Comparison Project [Starr et al., 2000]. Nucleation and ice crystal growth were forced through an externally imposed rate of lift and consequent adiabatic cooling (Table 1). The background haze particles are assumed to be lognormally-distributed $H_2SO_4$ particles. Only the homogeneous nucleation mode is allowed to form ice crystals in the HN-ONLY runs; all nucleation modes are switched on in the ALL-MODE runs. Participants were asked to run the HN-$\lambda$-fixed runs by setting $\lambda = 2$ ($\lambda$ is further discussed in section 2) or tailoring the nucleation rate calculation in agreement with $\lambda = 2^1$. The depth of parcel lift (800 m) was set to assure that parcels underwent complete transition through the nucleation regime to a stage of approximate equilibrium between ice mass growth and vapor supplied by the specified updrafts.

2 MODEL DESCRIPTIONS

Five parcel modeling groups participated in the CPMC (Table 2). Hereafter, we will refer to these models as the C, D, J, L, and S models, respectively, as denoted in the table.

The estimate of the nucleation rate of ice in solution droplets, $J_{\text{haze}}$, remains an active research area. $J_{\text{haze}}$ was computed using either (1) the modified classical theory approach (model J) or (2) the effective freezing temperature approach (hereafter, $T_{\text{eff}}$ models, models C, D, L, S).

The $T_{\text{eff}}$ models attempt to directly link measured $J_{\text{haze}}$ to nucleation rates of equivalent-sized pure water droplets $J_{w}$ via the effective freezing temperature, which is defined as

$$T_{\text{eff}} = T + \lambda \Delta T_m,$$  \hspace{1cm} (1)

such that $J_{\text{haze}} = J_{w}(T_{\text{eff}})$ as introduced by Sassen and Dodd [1988]. In (1), $\Delta T_m$ is the equilibrium melting point depression (positive valued), which depends on solute wt%, and $\lambda$ is an empirical coefficient to account for additional suppression/enhancement of nucleation temperature due to

$^1$Note that $\lambda = 2$ agrees approximately with data presented by Koop et al. [1998].
Table 2: Participant cirrus parcel models.

<table>
<thead>
<tr>
<th>Organization</th>
<th>Investigator</th>
<th>UKMO</th>
<th>CSU</th>
<th>ARC</th>
<th>GSFC</th>
<th>U. Utah</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cotton (C)</td>
<td>DeMott (D)</td>
<td>Jansen (J)</td>
<td>Lin (L)</td>
<td>Sassen (S)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bin characteristic</td>
<td>discrete</td>
<td>continuous</td>
<td>continuous</td>
<td>continuous</td>
<td>particle tracing</td>
<td></td>
</tr>
<tr>
<td>Haze size</td>
<td>$r_{eq}$ or $\frac{dr}{dt}$</td>
<td>$r_{eq}$</td>
<td>$r_{eq}$</td>
<td>$r_{eq}$</td>
<td>$r_{eq}$ or $\frac{dr}{dt}$</td>
<td></td>
</tr>
<tr>
<td>$\lambda$</td>
<td>1.5</td>
<td>1.5</td>
<td>varying$^c$</td>
<td>1.0</td>
<td>1.7</td>
<td></td>
</tr>
<tr>
<td>deposition coef. $\beta_i$</td>
<td>0.24</td>
<td>0.04</td>
<td>1</td>
<td>0.1</td>
<td>0.36</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Discrete vs continuous binning indicates if assuming that all particles have exactly the same size in a given size bin or a certain distribution of particle sizes is allowed in a bin.

$^b r_{eq}$ vs. $\frac{dr}{dt}$ denotes either using the equilibrium-sized haze approximation or computing the diffusional growth of haze particles explicitly.

$^c$ See section 2 for detailed discussion.

In model J, recent direct data on ice/solution surface tension was incorporated and activation energy was inferred from recent laboratory measurements of $J_{haze}$ for $H_2SO_4$ particles following Tabazadeh et al. [1997] and Koop et al. [1998]. This approach to determine $J_{haze}$ can be interpreted as a $T_{eff}$ scheme with varying $\lambda$ (Figure 1). The intrinsic $\lambda$ varies inversely with solute wt% and temperature. Also, the differences in the sensitivity of $J_{haze} V$ (V is the volume of the particle) to solute wt% between these two approaches may lead to systematic differences in the freezing haze size distributions. Nucleation rate data over a wide range of values, e.g., data points beyond critical freezing conditions, are needed to diminish the inconsistency between the two approaches.

Little constraint was imposed on formulating heterogeneous nucleation because theoretical and experimental understanding are still quite poor. Models C and L employ ice saturation ratio dependent parameterizations of activated IN following Spice et al. [1999] and Meyers et al. [1992], respectively. These parameterizations are expected to represent a maximum heterogeneous nucleation impact.

Haze particles of the given $H_2SO_4$ aerosol distribution are subject simultaneously to heterogeneous and homogeneous nucleation in models D and S. The number concentration of the activated IN in model D is computed following DeMott et al. [1998] based on field experiment data. This treatment was expected to yield the most conservative estimate of IN in cirrus. Model S computes the activated freezing nuclei using $T_{eff}$ dependent Fletcher equation [Sassen and Benson, 2000], where parameters were set to yield the most favorable conditions for heterogeneous nucleation.

Participants either assumed that haze particles are in equilibrium with the environment or computed the diffusional growth of haze particles directly (Table 2). The diffusional growth rate of haze particles more or less exponentially decreases with temperature as caused by water vapor saturation pressure. The response time scale to the deviation from equilibrium can be considerably greater than one model time step in a swift updraft in a cold environment. Therefore, large haze particles may

Figure 1: $J_{haze} V$ vs. temperature for solute wt% 5, 15 and 25%. Solid, dashed, dash-dotted, dotted curves denote models J, C, S, models D and L (same curves), respectively, for $\lambda = 2$. 

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non-ideal interaction between ions and condensed water. Although Sassen and Dodd [1988] noted that an average $\lambda$ for different solutions was around 1.7, values for specific solutions may range from 1 to 2.5.
become more concentrated than the corresponding equilibrium-size particles in such conditions. This may result in considerable delaying of haze growth in models C and S (Table 2) and affect ice particle formation rate.

3 RESULTS AND DISCUSSIONS

As we proceed to describe the results and differences between models, it must be noted that the benchmark is not necessarily the median or the average of model results. The predicted \( N_i \) (ice number concentration) at 800 m above the starting point is compared (Fig. 2). In the HN-ONLY cases, to a first order approximation, the logarithm of \( N_i \) increases quasi-linearly with the logarithm of updraft speed. The predicted \( N_i \) by models D, S and L are close; \( N_i \) by models J and C form the lowest and highest bounds in the six cases, respectively.

The cold cases

- \( W = 0.04 \) W = (0.21)
- \( W = 1.04 \) W = (0.21)

The warm cases

- \( W = 0.04 \) W = (0.21)
- \( W = 1.04 \) W = (0.21)

Figure 2: \( N_i \) predicted vs imposed updraft speed. The unfilled and filled bars denote HN-ONLY and ALL-MODE, respectively.

The cold cases

- \( N_i = 1 \) liter \(^{-1}\)

The warm cases

- \( N_i = 1 \) liter \(^{-1}\)

Figure 3: The \( RH_i \) at cloud base \( z_b \) and the corresponding \( \Delta RH_i \), defined as the difference between peak \( RH_i \) and \( RH_i \) at \( z_b \) (the HN-ONLY cases).

Figure 4: Ice water content (IWC), \( N_i \), ice particle formation rate \( \frac{dN_i}{dt} \), and \( RH_w \) as functions of \( z - z_b \).

Cirrus initiation occurred over a narrower range of altitude and \( RH_i \) (relative humidity over ice) in the warm HN-ONLY cases than in the cold cases (Fig. 3). The increasing sensitivity of the cloud base \( RH_i \) as temperature decreases in the four \( T_{eff} \) models is primarily caused by \( \lambda \).

Heterogeneous nucleation is a possible explanation of the discrepancy between the observed threshold \( RH_w \) for cirrus formation and the theoretically derived threshold \( RH_w \) (relative humidity over water) for homogeneous nucleation of \( H_2SO_4 \) or \((NH_4)_2SO_4 \) solution particles; e.g., [Heymsfield and Miloshevich, 1995]. Cirrus properties are affected by the dominant nucleation mode in cloud initiation because of the distinct characteristics of the two modes.

The cloud base height, \( RH_i \) and peak \( RH_i \) in the ALL-MODE cases (not shown) vary even more because of our respective unbounded choices of heterogeneous nucleation. The impact of heterogeneous nucleation on lowering \( N_i \), peak \( RH_i \), and cloud formation altitude is extremely sensitive to the onset conditions for nucleation and the subsequent ice particle formation rate. With heterogeneous nucleation, the peak \( RH_i \) is lower in all but the case Wa100 by model S. The predicted \( N_i \) is reduced in all but the case Ca004 by model S.

We now discuss the results of the HN-\( \lambda \)-fixed simulations. The nucleation regimes of Wh020L and Ch020L take place within the temperature range of -43.2 to -44.2°C and -63.2 to -64.2°C, respectively. The effect of temperature variation on nucleation rates within this 1°C range is secondary, compared to the evolution of haze solute wt%. Thus, it is justified to analyze and visualize results according to the \( z - z_b \) coordinate (Fig. 4).
in comparison to 3% and 8% in Wh020 and Ch020. The predicted \( N_i \) is only marginally affected.

At the beginning of the nucleation stage in Wh020L, ice particle formation rates by the four \( T_{eff} \) models are close. However, models C and D reach much larger \( RH_{w} \) that leads to larger instantaneous nucleation rates, and maintain the peak ice formation rate longer than the other two models.

Quite contrarily, the \( N_i \) curves of models D and L in Ch020L distinctly separate from those of models C and S. This grouping incidentally coincides with the grouping according to the haze size specifications. Large haze particles are more concentrated than the corresponding equilibrium values in models C and S. Yet, the nucleation regime in model S was not sustained as long as in model C; a similar finding is noted when comparing results of model D and L. The results of model J feature slow ice particle formation rate, long nucleation duration, and broader freezing haze number distribution.

The above results indicate that nucleation duration time and the maximum nucleation rate achieved are the two key components in determining the final \( N_i \). These two factors are sensitive to the growth rates of small ice crystals, which under the influence of the kinetic effect are sensitive to the deposition coefficient, \( \beta_i \). It was found that varying \( \beta_i \) from 0.04 to 1 (Table 2) would result in a factor of 4-5 (Wh020L) and 9-12 (Ch020L) variation in \( N_i \) by models C and L.

4 SUMMARY

Results of Phase 1 of CPMC projects show that the predicted cloud properties strongly depend on updraft speed. Significant differences are found in the predicted \( N_i \). Detailed examination revealed that the homogeneous nucleation formulation, aerosol size specification, ice crystal growth (especially the specification of the deposition coefficient for ice) and water vapor uptake rate were the critical components. These results highlight the need for new laboratory and field measurements to infer the correct values for critical quantities in the cirrus regime.

No attempt was made to scrutinize the causes of differences in ALL-MODE simulations due to the substantial differences in formulation of heterogeneous nucleation. Nevertheless, it was confirmed that the expected effect of a heterogeneous nucleation process is to decrease \( N_i \) and the \( RH_{w} \) required for cloud initiation. Clearly, new measurements of ice nuclei activation in cirrus conditions are warranted.

CPMC Phase 1 was conducted based on a single CCN distribution. Phase 2 of the CPMC, now underway, examines the effects of varying aerosol distributions. Sensitivity of model results to CCN composition is indirectly made by altering \( \lambda \).

5 ACKNOWLEDGMENT

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6 REFERENCES


