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Interactive Comment

Interactive comment on "Simulating mixed-phase Arctic stratus clouds: sensitivity to ice initiation mechanisms" *by* I. Sednev et al.

I. Sednev et al.

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Response to Reviewer #3

We thank the reviewer for the many comments/suggestions made. We have addressed the questions and concerns of the reviewer and will modify the paper accordingly. Our response to the questions raised are as follows:

"The authors are conducting sensitivity modeling studies, based on MPACE observations, assuming two different types of ice initiation processes. For the nucleation process of ice crystals from water vapor the Meyer's formula is used to calculate the number of ice crystals. On page 11761, line 14, it is state that "if liquid phase is not involved, ice initiation is parameterized with the Meyers formula" (i.e freezing mode must therefore be deposition freezing). However, it should be noted that the Meyer's formula was developed based on measurements with a Continuous Flow Diffusion Chamber (CFDC) that measures freezing from both deposition and condensation freezing (freezing where CCN that has activated a droplet also serve as an ice nuclei (IN), i.e. from liquid phase). Therefore, when the Meyer's formula is used as it is, more than one basic ice initiation process is accounted for. The explicit ice freezing modes that the Meyer's formula is based on (deposition and condensation) should therefore be stated in the paper."

Heterogeneous condensation freezing does not require the presence of supercooled water droplets. In this process "liquid water" forms on the ice nucleus followed by immediate freezing. The difference between immersion freezing and condensation freezing is based on relative amount of water present and time during which the "liquid"; exists. Meyers formulation does not include liquid phase and deals only with water vapor. At the same time it really accounts for two processes. We use generic term for the process of initiation of ice phase from water vapor and write on page 11761: If liquid phase is not involved in ice initiation process, we parameterize nucleation of ice crystals from water vapor as a function of SSI (Meyers et al., 1992):

Nmc = Nms x exp[Ams + Bms x Si] (1)

where Ams and Bms are set to -0.639 and 12.96, respectively, Nms is ice nuclei (IN) concentration in 1/L, and Nmc determines the upper limit of concentration, up to which ice crystals can be nucleated from water vapor at a particular point.

"Another issue is that IN measurements, with the same type of CFDC as used to develop the Meyer's formula, was conducted during the MPACE study [Prenni et al., 2007]. Prenni et al., determined that the Meyer's formula overpredict IN concentrations by a factor of 26. Note that the Meyer's formula is developed from measurements at midlatitudes levels where the concentration of dust (one of the main contributors to the IN population) typically is higher than at Arctic latitudes. Prenni et al. also found that measured IN concentrations during MPACE had a smaller temperature dependence

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than is in the Meyer's formulation. Hence they developed a new formulation, but with the same functional form as the Meyer's equation. And, again, this formula is for both condensation and deposition mode freezing. I suggest including a reference to Prenni et al. and a small discussion on how the sensitivity study might compare with the IN measurements. Even, if possible, use the parameterization developed from the specific MPACE measurements."

This is not an issue. We are aware how Meyers formulation was derived as well as environmental ranges which it can be applied for. Prenni et al. (2007) (P2007) proposed non-temperature dependent formula that has the same functional form as formula (2.4) in Meyers et al. (1992) (M1992), which we use to parameterize nucleation of ice crystals from water vapor (our formula (1)). Values for Nms, Ams, and Bms used in P2007 are 1.0 1/L, -1.488, and 0.0187, respectively. In our sensitivity experiments we use different values for Nms (Table 2). For example, these value are 1.0 1/L and 0.1 1/L for runs I1 and I2, respectively. Nmc values calculated using our formula (1) in I2 and P2007 formula coincide (0.285 1/L and 0.288 1/L, respectively) when supersaturation w.r.t. ice (SSI) is about 13 %. For SSI between 2%-13% and 13%-25% these two formulae provide mean values equal to 0.154 1/L and 0.260 1/L and 0.663 1/L and 0.320 1/L, respectively. Mean Nms ratios for our formula (1) as used in I2 and P2007 formulation are 0.591, 2.071, and 1.506 for SSI between 2%-13% , 13%-25%, and 2%-25%, respectively. Both formulae give Nmc that has the same order of magnitude for wide SSI range if Nms is set equal to 0.1 1/L in our formula (1). It can be demonstrated on P2007 Fig. 3 by parallel shifting of blue line that depicted M1992 formulation. Nms is set equal to 0.1 1/L in our sensitivity runs I2 and I4 (Table 2). If formula (1) were replaced by P2007 formulation in these runs, we would expect increasing significance of the second IIP. We would not expect to get dramatical changes if P2007 formulation were used. The relative significance of expected differences is determined by how often simulated SSI is less than 13 % (P2007 formulation overpredicts observed MPACE values) and greater than 13 % (our formulation overpredicts observed MPACE values). In this paper we use in formula (1) exactly the same values for Ams and Bms as in

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M1992. In opposite to P2007 who changed Ams and Bms values in M1992 formula we changed Nms value (see discussion above). By doing so we clearly recognize how M1992 formula was developed as well as environmental ranges which it can be applied for. Moreover, Nms that is constant in this study can be a function of altitude and geographical location (among others) if used in global models. In this case it would be better to treat Ams and Bms as constants and make changes to Nms in a manner that it is done to account for maritime and continental CCN concentration differences in some GCMs.

"Further, some of the basis for the main conclusions in the paper is not described well enough (see specific comments below)."

SPECIFIC COMMENTS:

"I would suggest using a different word for newborn droplets (for example newly activated)"

We agree. "newly activated" or"just nucleated" are equally good.

"Page 11759, line 20: I suggest using the term deposition/condensation freezing when describing the ice initiation process from water vapor."

We prefer not to use deposition/condensation freezing for IIP when the presence of liquid phase is not required. We only use the empirical relation that shows dependence of IN on SSI and do not suggest what the real physical processes that determine its existence are (probably ionization and ions as IN are important). In our paper we use generic terms "nucleation from water vapor" for the first IIP and "transformation of super-cooled liquid water" for the second IIP.

"Page 11761, line 13: Suggest changing "...some of which (IIP and BFP) are of special .." to "...for where IIP and BFP are of special ....""

Our sentence is as follows:

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The BRM scheme takes into account numerous microphysical processes, some of which (IIP and BFP) are of special interest in this study.

We could not follow this suggestion. We prefer to keep our sentence as is.

"References related to Equation (2) should be included in the main text and not only in the appendix. I also suggest that the discussion of ice initiation mechanisms in the appendix could be moved to the main text, since this aspect is one of the main focuses in the paper. "

The text will be modified.

"The effective radius definitions in appendix could also be moved to the main text."

It will be moved to the main text if we decide to drop our appendix.

"Page 11763: A_t, B_t, C_t and P_t have wrong subscripts. It should be "l" and not "t"."

We agree.

"Page 11763: It is more likely that the second mode of the AP distribution (the larger sizes with median radius of about 1 micrometer) would be comprised of dust/soil like particles, and not only pure ammonium sulfate. The particles in this mode (if assumed to be dust) can still act as CCN though [e.g. Mahowald and Kiehl, 2003], since the sizes are large (giant CCN). In addition, Koehler et al. [2007] show that dust with only slight soluble material on them is hygroscopic and can serve as CCN. This can later then also lead to condensation freezing. "

Broadness of CCN spectra is important for the second IIP. It is obvious that in hypothetical situation when chemical composition of the second AP mode is different from that of the first mode, broadness of just nucleated droplet spectra and relative significance of the second IIP might be affected. In this paper we don't focus on how AP of different chemical compositions influence the IIP mainly since the observation were

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not available , though the BRM scheme is designed to account for this.

"Page 11766, line 13: Can you explain why precipitation flux is lower and droplet concentration is higher in the W3 case compared to W4. Intuitive, I would think precipitation would increase with coagulation and droplet concentration would decrease with coagulation."

Definition of precipitation flux is given on page 11781. Increase and decrease in concentration don't guarantee that changes of precipitation flux occur because of its dependences on the bin mass and terminal velocity. Coagulation reduces concentration and increases precipitation flux. At the same time reduced concentration might lead to supersaturation increase, and numerous small droplets could be nucleated. In our simulations this non-linear dependence is determined to a great extent by implied forcing and prescribed AP distribution.

"Page 11767, paragraph starting on line 3: Statements in this paragraph are not well founded based on the information given in the paper. It is not clearly shown or discussed why and how the CCN spectrum shape is more important than the process of coagulation. Simulations with a different (monomodal or less broad?) distribution could maybe support this statement? "

We discussed this in "warm" sensitivity run Section. We imply that the reader has a basic understanding regarding parameterization of droplet nucleation process in BRM schemes. We briefly mentioned this scheme on page 11761 (line 9) and page 11764 (line 24). We are not sure that more detailed description should be included.

In the BRM scheme the parameterization of the activation of aerosol particles to form cloud droplets depends on the assumed aerosol composition and supersaturation w.r.t. water. In our runs aerosol particles of a certain size are activated when the supersaturation calculated exceeds the critical value determined by the Koehler equation assuming an ammonium sulfate composition. Koehler theory is used to determine so called critical supersaturation and critical and equilibrium radii. In the case the AP distribution

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contains aerosols with dry radius greater than critical radius at a given point, these APs can be activated and transformed into droplets. The size of new nucleated droplets is equal to equilibrium radius if dry AP radii is less than 0.03 microns; otherwise, the radii of cloud droplets are five times as much as dry AP radii since large CCN does not reach their equilibrium sizes. This approach prevents origination of unrealistically large droplets and too fast warm rain formation (Kogan, 1991; Khain and Sednev, 1996). This droplet nucleation scheme starts with activation of the APs, whose critical supersaturation is the smallest, calculates corresponding droplet sizes and liquid content increase, and assures that total vapor and cloud water content is conserved and there is still enough water vapor to activate APs in the next bin. If the coarse AP mode is present, it is obvious that treatment of droplet nucleation outlined above will produce broader droplet distribution and increase amount of relative big droplets whose freezing probability is greater than that of smaller droplets as given by equation (2) in our paper. To emphasize this fact additional runs are not needed. Because this approach for droplet nucleation and its modification (Yin et el., 2004) is now routinely used in different BRM schemes, we decided not to focus on it.

"In addition, the next sentence is somewhat out of place. It is new for the reader that it is assumed that "water-water, ice-water and ice-ice interactions may be relatively minor for the MPACE single-layer mixed phased clouds." No simulations showing ice-water and ice-ice interactions are shown yet, thus I cannot see how only the warm cloud simulations can validate the above mentioned assumption. This also brings up a new question. Is coagulation included in the "ice" microphysics sensitivity runs? This is not clearly stated in the paper."

We perform our "warm" sensitivity runs to ensure that simulated cloud characteristics for liquid phase (droplet concentration, content, and effective radius) are in range with observations. In addition, these runs permit us to evaluate influence of initial setup (temperature, water vapor mixing ratio, and microphysical characteristics vertical profiles and bimodal distribution of dry aerosols) and applied forcing (temperature and

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water vapor mixing ratio tendencies, prescribed subsidence and surface sensible and latent fluxes) proposed for MPACE intercomparison study (Klein et al., 2007) on simulated clouds. Based on numerous supplemental runs (not shown) and analysis of observation we conclude that water-water interactions (coagulation) is relatively minor if we believe that applied forcing and AP distribution are typical for MPACE period B conditions. Moreover, because of small concentration of ice particles and significantly reduced collision efficiencies for water-ice and ice-ice interactions at low temperatures, we conclude that coagulation is relative unimportant as compared to Bergeron-Findeisen process. That is why coagulation is not included in the "ice" microphysics sensitivity runs.

"Page 11769, line 8: It is not clear for me, looking at Figure 12 that I3 predict significant higher crystal concentrations than I2. I suggest reducing the maximum range in color table to better illustrate differences in Figure 12 (and Figure 11)."

We agree. The color bar should be changed in some figures. Table 7 can be used to see a difference.

"Discussion on page 11773 and 11774: It is stated that the second ice initiation process "crucially depends on the shape of the AP distribution and not only on the concentration of cloud droplets". "

"First, it is not clearly shown how the broadness of the AP distribution affects the ice initiation process compared to a lesser broad AP distribution."

If droplet nucleation occurs, the coarse mode of AP distribution is an instant source of relative big droplets (more then 10 microns). In accordance with our formula (2) supercooled cloud water transformation rate in each bin depends on the mass of this bin (among others). Please, distinguish between "mass of bin" and "mass in bin". Assuming mono-disperse distribution and the same concentration for cloud droplets, this rate differs by more than two orders of magnitude for 2 micron and 16 micron droplets (increase in droplet mass is equivalent to decrease in time that is necessary to re-

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duce droplet concentration by half). The broadness of AP spectrum determines the broadness of spectrum of cloud droplet just nucleated and consequently the second IIP efficiency for both saturated and subsaturated (w.r..t. water) environment. If SSW $>$ 0 transformation of liquid phase into ice phase is accompanied with droplet condensational and ice depositional growth; otherwise, the same transformation occurs when droplets are evaporated supplying additional water vapor for ice particle depositional growth (Bergeron-Findeisen process). Even if liquid particles evaporate and ice particles sublimate, this transformation takes place. Mainly due to implied forcing (SSW $<$ 0) simulated clouds develop in subsaturated (w.r.t. water) environment as is mentioned in our paper. If supersaturation w.r.t. water occurs, cloud droplets with radii more than 10 microns are nucleated. Evaporation of droplets is accompanied with freezing. These environmental conditions are not favorable for coagulation, preventing droplets growth up to the sizes where their collision efficiencies become significant.

"Second, what about IN concentrations in the droplets? For a droplet to freeze, an IN must be present. A larger droplet will have a larger freezing probability due to higher concentration of IN in the droplet (could be acquired for example through coagulation with droplets containing IN, or scavenging of IN). Would there be a difference in freezing probability from a large droplet originating from a broad CCN distribution, or a large droplet grown due to coagulation?"

To account for effect mentioned by reviewer two-dimensional size distributions functions are needed (Bott, 2000). In this approach the particles are classified according to their water and total aerosol mass on a two-dimensional grid. For example, Diehl and Wurzler (2004) studied heterogeneous drop freezing employing the two-dimensional treatment of cloud physics that allows the coexistence of similarly sized drops with different contents of soluble and insoluble particles. In our simulations the one-dimensional approach is used, and freezing probability of equally sized droplets remains the same and does not depend on origination mechanism. We should note that the two-dimensional treatment of mixed-phase cloud microphysics is very compu-

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tationally expensive even in parcel model simulations.

"Page 11779, line 8 and 9: What is the difference in meaning of "activation of CCN", and "nucleation of droplets"?"

We don't resolve explicitly the process of AP condensational growth and process of droplet formation, but we parameterize them. In these parameterizations:

Activation of CCN means calculation of so called "critical radius of dry AP" above which AP can be served as CCN.

Nucleation of droplets means calculation of spectrum of new cloud droplets.

"There should be more references to newer studies of ice initiation processes. There has been a lot of work on this the last 14 years (since Deshler and Vali, 1992). Among some of the newer ones are for example DeMott et al. [2003], Archuleta et al. [2005], Mangold et al. [2005], Field et al. [2006], Mohler et al. [2006], Koehler et al. [2007] and Marcolli et al. [2007]."

Thanks for providing us with these references. After reviewing references, we decided to include some of these and other references. For example, paper by Diehl and Wurzler (2004) who consider the significantly different ice nucleating efficiencies of various ice nuclei is referenced.

"Page 11781, line 18: Two main mechanisms for ice generation by supercooled droplets are mentioned. What about condensation freezing? (see Vali, [1985])"

Heterogeneous condensation freezing does not require the presence of supercooled water droplets (Young, 1974; Fukuta and Schaller, 1982).

FIGURES AND TABLES:

"Figure 11 and 12: I suggest reducing the maximum range in color table to better illustrate differences in the figures."

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We agree.

"Table 8: In table heading, include equation number. In table caption, use correct subscript for plates and dendrites."

Equation numbers are included in table caption.

" TECHNICAL COMMENTS:

"simplier" on page 11758, line 12 should be spelled simpler

Page 11767, line 13, CN should be CCN.

Page 11777, line 10, an "l" is missing in complexity.

Page 11785, "Combining" is spelled wrong.

Use a different word for newborn (for example newly activated....) Acronyms are defined unnecessary multiple times. Replace all occurrences of undersaturated with subsaturated"

Thanks, all these are accepted. We will modify our text accordingly.

References:

Fukuta, N. and Schaller R.C.: Ice nucleation by aerosol particles: Theory of condensation-freezing nucleation. J. Atmos. Sci., 39, 648-655, 1982.

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