Interactive comment on "Aerosol and dynamic effects on the formation and evolution of pyro-clouds" by D. Chang et al.
MS No.: acp-2014-61

Dear Reviewer,
We would like to thank you for the valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below (line numbers refer to the new version of manuscript). In addition, the title of the manuscript is revised to be "Regime dependence of aerosol effects on the formation and evolution of pyro-convective clouds".

Reply to Anonymous Referee #1

This paper is a follow-on study on aerosol effects under different heat forcing conditions by conducting 2-D simulations using the Active Tracer High Resolution Atmospheric Model with a two-moment cloud microphysics parameterization. Although the different sensitivity regimes are classified and associated processes are analyzed, the results of the study are not new (some results have been the common senses for scientists in this area) and many previous studies with even more advanced microphysics have indicated similar results. Since most of those previous studies are 2-D, 2-D dynamics is very different from 3-D, and this study has an emphasis on dynamic effect, investigations with 3-D simulations would be something building on past research.

The paper does not provide further explanations for the phenomena they see (see specific comments). The introduction of the paper is poorly written (see details in my specific comments). Most importantly, many process rates heavily depend on the process parameterizations (scheme-dependent), but there is no any discussion about those uncertainties.

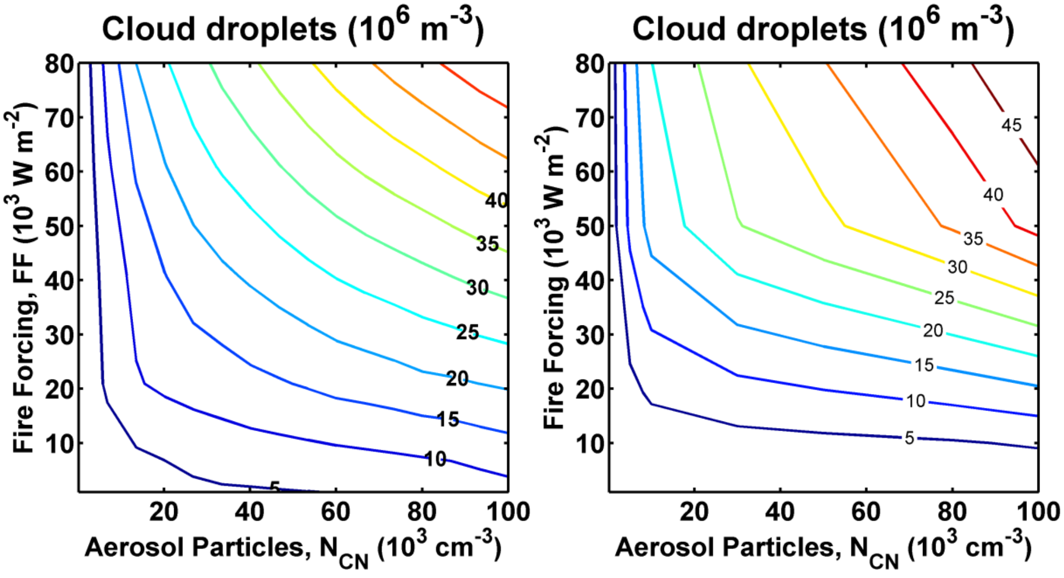
The paper is misleading in wording such as fire forcing and biomass burning aerosols.

What I found out eventually is that there is nothing to do that fire and biomass burning aerosols. It is just a heat forcing to produce different intensity of updrafts. See my specific comments for details.

Therefore, the paper needs very significant revisions to reach the point being accepted as a publication.

Response: Thanks for the comments. We have revised the introduction part with an emphasis on clarifying the motivations and new aspects of this work. One motivation is to determine the regime-dependence of aerosol effect. The other problem that we try to solve is the nonlinearity in the aerosol-cloud interactions. In the main text, more discussions concerning the uncertainties of microphysical scheme have been added.

Besides, the results and discussions from the 3-D simulations have been included in the supplementary material. Take cloud droplets for example (Fig. R1), the regime dependence from the 3-D simulations (Fig. R1b) looks similar to the 2-D results (Fig. R1a) though the absolute dependency may vary. This suggests that the use of such regime dependence requires caveats because it may differ for different model dimensionality (2D vs. 3D). In the main text, we have included more discussion concerning these uncertainties: "In this study, we demonstrate the performance of ensemble simulations in determining the regime dependence of aerosol effects. The use of such regime dependence requires caveats because it may differ for different cloud types, aerosol properties, meteorological conditions and model configurations (e.g., microphysical schemes, dynamic schemes, dimensionality, etc.; the 3-D results are in the supplementary material)". Please see Lines 739-743.



(a) (b)

Figure R1. Number concentration of cloud droplets calculated as a function of aerosol number concentration (N_{CN}) and updraft velocity (represented by FF) from 2-D (a) and 3-D (b) simulations.

The wording "fire forcing" was adopted from previous application of ATHAM model (Luderer, 2007) for the same fire event. The wording "biomass burning aerosols" is used because the CCN activation properties in our model simulation are taken from that of biomass burning aerosols.

Specific comments

1. Introduction 1. First paragraph of introduction needs to be cleared up a lot. First, aerosol impacts on precipitation are very different for different cloud types such as shallow warm clouds and deep convective clouds. Therefore, not to be more confusing, please discuss them by separating cloud types.

Response: Thanks for the suggestion. We discuss the aerosol effects on the precipitation formed in different cloud types separately, mostly stratiform and convective clouds. The text is "Precipitation from stratiform clouds can be inhibited by elevated aerosol concentration (Zhang et al., 2006), while precipitation from convective clouds can be either suppressed or enhanced (Ackerman et al., 2003; Andreae et al., 2004; Altaratz et al., 2008; Lee et al., 2008; Teller and Levin, 2008; Fan et al., 2013; Camponogara et al., 2014)." Please see Lines 51-54 in the new manuscript.

2. P7779, Line 18-21: Li et al. 2008 detailed the non-monotonic behavior. Line 21-25: Qian et al. 2009 was not such study. Khain 2009 and Fan et al. 2009 are typical such studies.

Response: The sentence in Line 18-19 in old manuscript is revised to be "In addition, changing aerosol concentrations have also been found to exert non-monotonic influences (either positive or negative) on a wide range of cloud properties". Please see 54-56 in revised manuscript.

The work of *Khain (2009) and Fan et al. (2009)* indicated that the environmental conditions (e.g., relative humidity, wind shear) could regulate the aerosol effect on the cloud and precipitation. Here we have deleted the citation of *Qian et al. (2009)* paper, and add the citation of *Khain (2009) and Fan et al. (2009)* paper. Please see the revised sentence in Lines 59-65 in the new manuscript: "One explanation for these seemingly contradictory results is that aerosol effects are regime-dependent, which means that it can vary under different meteorological conditions (updraft velocity, relative humidity, surface temperature, and wind shear), cloud types, aerosol properties (size distribution and chemical composition) and observational or analysis scales (Levin and Cotton, 2007; Tao et al., 2007; Khain et al., 2008; Rosenfeld et al., 2008; Fan et al., 2009; Khain, 2009; Reutter et al., 2009; McComiskey and Feingold, 2012; Tao et al., 2012)."

3. Recent progresses on aerosol effects on convective clouds are not introduced. For example, a recent review study (Tao et al., Rev Geophys, 2012) on aerosol impacts on convective clouds is not even mentioned. A nice related paper on the relative importance of the thermodynamic and microphysical aerosol effects (Fan et al., PNAS, 2013) is missed too. Anyway, there are so many significant studies on aerosol impacts on convective clouds since 2011 in literature (Morrison, van den Heever, etc) but these progresses are not discussed at all. It is recommended that the authors do a thorough literature study of this topic.

Response: Thanks for the comments. Tao et al. (2012) summarized the aerosol effects on the CCN activation, warm-rain process, mixed-phase clouds, and precipitation in terms of microphysical scale, cloud-resolving scale, and regional scale, which are retrieved from the theoretical analysis, observations, and numerical modeling. The underlying mechanisms and the comparison between the results from different studies was also presented and analyzed. Fan et al. (2013) carried out monthly 3-D simulations over three different regions and found the microphysical effect controlled by aerosols is the major factor that determines the properties of deep convective clouds, rather than the updraft-related dynamics. The introduction has been reformulated with these suggested papers which will be included in the revised manuscript.

4. The third paragraph of introduction: I do not see how your study is connected with biomass burning aerosols, only through the heat you added? Are the aerosol properties used in the study taken from biomass burning aerosols? If not, this is just a general test, not for biomass burning aerosols.

Response: We connect our study with biomass burning aerosols through the setting of aerosol properties. The properties of aerosol particles (used in the look-up table for cloud nucleation process) are for soot particles. In the revised manuscript, we emphasize the connection between our simulations and biomass burning aerosols in Sect. 2.2. The corresponding text is "As mentioned above, we used the lookup table of Reutter et al. (2009) for the CCN activation. This table is determined for fresh biomass burning aerosols with a hygroscopicity parameter κ of 0.2 and a log-normal size distribution (a geometric mean diameter of 120 nm and a geometric standard deviation of 1.5, Reutter et al. 2009)." Please see Lines 157-160.

5. The motivation based on Reutter et al 2009 (as stated in the first sentence of the abstract) is missing from the introduction.

Response: In the third paragraph of the introduction section, we extend the motivation stated in abstract in terms of the fact that aerosol effect on convective clouds is regime-dependent based on the research from Reutter et al. (2009) and some other previous studies. The corresponding text can be found in Lines 59-70: "One explanation for these seemingly contradictory results is that aerosol effects are regime-dependent, which means that it can vary under different meteorological conditions (updraft velocity, relative humidity, surface temperature, and wind shear), cloud types, aerosol properties (size distribution and chemical composition) and observational or analysis scales (Levin and Cotton, 2007; Tao et al., 2007; Khain et al., 2008; Rosenfeld et al., 2008; Fan et al., 2009; Khain, 2009; Reutter et al., 2009; McComiskey and Feingold, 2012; Tao et al., 2012). It is thus important to investigate the regime-dependence of aerosol-cloud interactions and to improve the representation of cloud regimes in models (Stevens and Feingold, 2009). If we were able to distinguish under which conditions cloud formation is updraft-limited (aerosol-insensitive) as discussed in Reutter et al. (2009), it would have the advantage that in future work one could for many purposes neglect aerosol effects on clouds in areas that are usually updraft limited."

6. Section 2.2: It is very confusing by saying fire forcing. I was misled by the wording and thought that a real fire situation is set up such as T, RH, and aerosol emissions from fires. Until I finished the whole section, I realized that it is not about fire forcing at all. It is just a heat forcing to produce different intensity of updrafts (if for fire, at least aerosol emissions from the heating plume should be assumed, not the uniform aerosols over the entire domain). Based on the general aerosol type and a simple heating setup, please remove all those fire forcing or biomass burning aerosols.

Response: Thanks for the comments. We adopted the terminology "fire forcing" from the paper of Luderer (2007) for the same fire event, which stated that the "fire forcing" results in the vertical development, and favors for the formation of pyro-convective clouds. Within our simulations, the variation in updraft velocities is through changing the input fire forcing.

We admit that we did not consider the spatial and temporal distributions of atmospheric aerosols during the simulation. Instead of this, the concentration of ambient aerosols is set to be homogeneous over the modeling domain. We admit the variability of aerosols during the simulation is ignored and may leads to a bias compared to a real fire (Wang et al., 2013). However, this study aims to estimate the sensitivity of clouds and precipitation to the orders-of-magnitude change in the aerosol concentrations, and the similar treatment of aerosols has been used in previous studies (Seifert et al., 2012; Reutter et al., 2013). We add this discussion about the bias in the revised manuscript; please see lines 152-156. This work follows the old terminology (fire forcing) and treatment of aerosols from previous research paper. For the future research, we will try to improve the representation of the aerosol particles and take into account the full complexity of all chemistry-aerosol-cloud interactions.

Section 2.3: The process analysis used here is not something new or unique. Modeling studies like this do those analyses all the time. I do not see why a section is needed to introduce the analysis. Simply, you only need 1-2 sentences to introduce the table A1 for the quantities you look at.

Response: Thanks for the comment. Within the ATHAM model, process analysis is a newly-developed module, and here we tried to state the development on the existing model scheme. Different from the simple process rate calculations under four extreme conditions in the previous version of this paper, in the revised manuscript, we figure out how each microphysical process contribute to each hydrometeor over a wide range of aerosol concentrations and fire forcing, just as shown in Fig. R2. What is the percentage of the contribution of each process and how do they response to the changing updrafts and aerosols in the atmosphere? This is the fundamental questions with which the newly-developed PA module tries to deal. In Sect. 2.3, we intended to make clear what we have changed inside the model and what we want to get from it. So far as we know, there was no similar form of process analysis of cloud microphysics in literature.

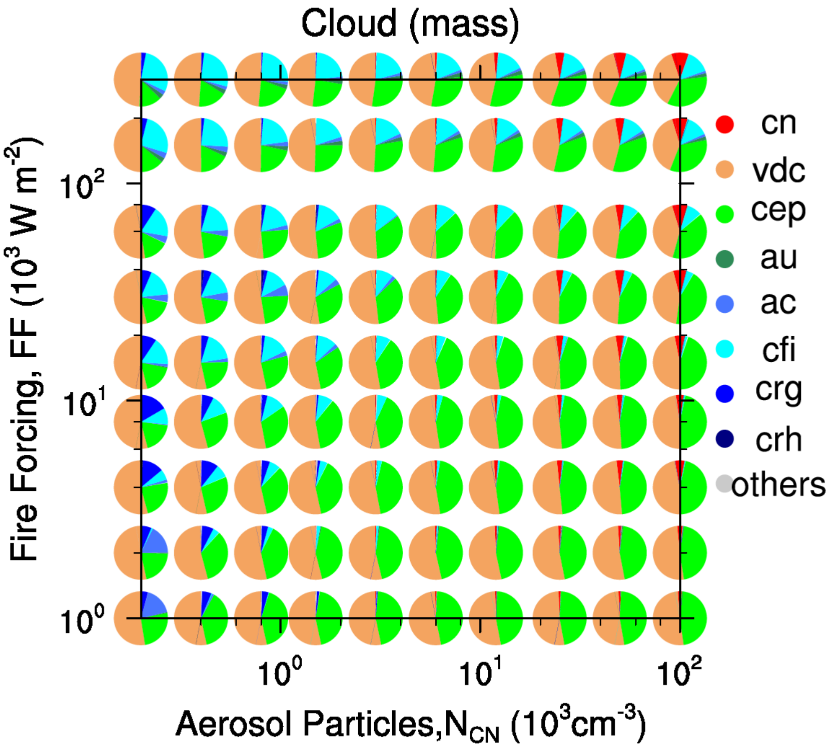


Figure R2. The pie charts summarize the relative percentage of the microphysical processes involving cloud droplets as a function of N_{CN} and fire forcing. Colors within each pie chart reflect the contribution of processes under the specific condition. Warm colors denote the source, while cold colors denote the sink. The acronyms indicate cn: cloud nucleation; vdc: condensational growth of cloud droplets; cep: evaporation of cloud droplets; au: autoconversion; ac: accretion; cfi: freezing of cloud droplets to form ice crystals, including homogeneous and heterogeneous nucleation; crg/h: riming of cloud droplets to form graupel/hail.

Section 3.2 Please use the temperature instead of Wm-2 to be more straightforward to general readers about the heating that you imposed in the experiments throughout the paper.

Response: Thanks for the suggestion. We plotted the relationship between fire forcing and the corresponding maximum temperature at cloud base under different aerosol conditions, and found the aerosol impact on the temperature is negligible. Take N_{CN}=5,000 cm^{-3} for example, the correlation of fire forcing and temperature is shown in Fig. R3. The shaded area indicates the variability of estimation over each simulation period. According to the figure, the temperature at cloud base varies monotonically from 7.6 to 16.4 ℃ as fire forcing increases from 1×10^3 to 3×10^5 W m^{-2}. We add this discussion in Sect. 3.1. Please see Lines 221-230.

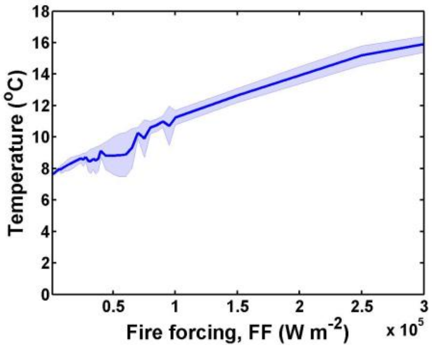


Figure R3. The correlation of fire forcing and the corresponding maximum temperature at cloud base. The shaded area indicates the variability of estimation (±½σ) over each simulation period.

In Sect. 3.2.1, we add the temperature as the secondary vertical axis in the contour plot for reference, as displayed in Fig. R4 (Fig. 7b in the revised manuscript).

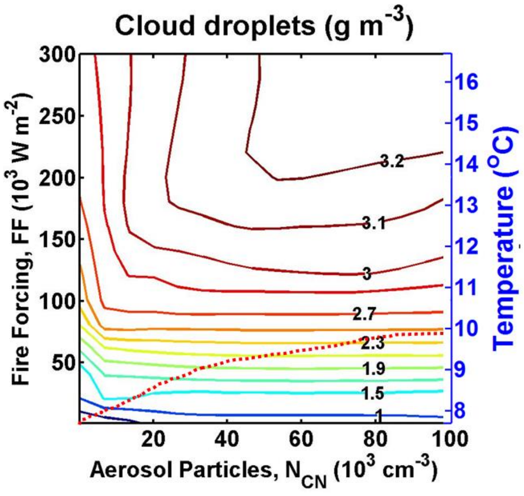


Figure R4. Mass concentration of cloud droplets calculated as a function of aerosol number concentration (N_{CN}) and updraft velocity (represented by FF). Red dashed lines indicate the borders between different regimes defined by $RS(N_{\mathrm{CN}})/RS(FF)=4$ or 1/4, respectively.

Section 3.2.1 Need to explain the reasons for aerosol-limited, updraft-limited and the transitional regime.

Response: For the three-regime structure of number concentration of cloud droplets in Sect. 3.2.1, we discussed the reasons in detail in Sect. 3.3.1. Please see Lines 426-450.

Section 3.2.2 The sensitivity of raindrop really depends on autoconversion parameterization, snow/graupel/hail productions and melting processes. All those parameterizations have very large uncertainties, especially with bulk microphysical parameterizations.
For example, most of the autoconversion schemes were developed or evaluated for stratocumuls clouds. They may not be appropriate for convective clouds. All I want to say is that the authors have to be aware of all these uncertainties and discuss them accordingly.

Response: Thanks for the suggestions. We explained the response of raindrops to aerosols/fire forcing (Sect. 3.2.2) in Sect. 3.3.2. Concerning the uncertainties of individual microphysical processes, we share the same concern as the referee. In our study, we have quantitatively shown the importance of different microphysical processes in regulating the number and mass flow of clouds. Currently, more efforts have been spent on improving the description of CCN/IN activation. However, the overall uncertainties will not be reduced if understanding the other microphysical processes is not improved as well.

Evaluating the uncertainties of all microphysical processes, however, will become a comprehensive review going beyond the purpose of the present study. In Sect. 3.3.2 of the revised manuscript, we add more discussion about the uncertainties concerning the main microphysical processes relevant to raindrops. The text is “The sensitivity of raindrops to aerosols depends on autoconversion parameterization, and the melting processes, etc. All those parameterizations have very large uncertainties, especially with bulk microphysical parameterizations. For example, most of the autoconversion schemes were developed or evaluated for stratocumulus clouds, which may not be appropriate for convective clouds. Based on the simulations during the convective phase of squall-line development, van Lier-Walqui et al. (2012) presented the uncertainty in the microphysical parameterization by the posterior probability density functions (PDFs) of parameters, observations, and microphysical processes. With the purpose to improve the representation of microphysics, it is of significance to quantify the parameterization uncertainty by using observation data to constrain parameterization." Please see Lines 547-556.

P7789, first paragraph, it is very vague by using buffering effect to explain the less sensitivity. Please stick on processes.

Response: Yes, by adding a sentence in lines 329-330 "Detailed analysis of the microphysical buffering processes will be presented in Sect. 3.3.2.", we will direct the readers to the process analysis. The text for the process analysis in Sect. 3.3.2 is "The PA clearly demonstrates that aerosols could significantly alter the microphysical pathways and their intensities. Although the variation in individual microphysical process is remarkable, the net result of all processes is not obvious and even insusceptible to aerosol perturbations. This is especially obvious when we consider the aerosol effect on rain water: it is observed that as aerosols is enhanced by a factor of 500, the intensities of the source processes only decrease by a factor of 10; however, there is only a two-fold change in the net rain water content. This implies that the microphysical scheme itself is a self-regulatory system, which can produce equilibrium and buffers the effect of aerosol disturbance (negative feedback)." Please see lines 538-546.

Section 3.2.3 Again for frozen water content and particle numbers, ice nucleation parameterizations and drop freezing parameterizations impact them dramatically. Please connect them with the parameterizations of these processes in your model and discuss the uncertainties.

Response: Thanks for the suggestions. We explained the response of frozen particles to aerosols/fire forcing (Sect. 3.2.3) in Sect. 3.3.3. In Sect. 3.3.3, we analyze the change trend of frozen particles in terms of microphysical processes, and try to find the dominant factors that regulate the process rate. As explained before, reviewing the uncertainties of all microphysical processes is beyond the current work. In Sect. 3.3.3 of the revised manuscript, we add more discussion about the possible uncertainties of these processes based on previous research. The text is “As shown aforementioned, drop freezing parameterizations and ice nucleation parameterizations influence frozen water content dramatically, which involve large uncertainties. Ice microphysics is significantly more complicated due to the wide variety of ice particle characteristics. On one hand, the intensities of these processes differ greatly among different microphysical schemes. Eidhammer et al. (2009) have compared three different ice nucleation parameterizations, and found that different assumptions could result in similar qualitative conclusions although with distinct absolute values. The parameterization with observational constraints agrees well with the measurements. On the other hand, van Lier-Walqui et al. (2012) suggested the processes contributing to frozen particles are dependent on both particle size distribution and density parameters. Parameterization improvement based on observations could help to reduce the uncertainties.” Please see Lines 597-607.

Section 3.2.4 Need to provide the reasons to explain the enhanced and suppressed rain rate regimes.

Response: We run more simulations and conduct more analysis to solve this question. By doing process analysis (PA), the most of rainfall is from melting of frozen particles. As shown in Fig. R5, the green diamond points is averaged rain rate under different aerosol concentrations. The columns represent the integrated melting rate from individual frozen particles. We found the rain rate is well correlated with the melting rate (as shown in Fig. R5). For $N_{\mathrm{CN}} > 1{,}000\ \mathrm{cm}^{-3}$, increasing N_{CN} results in more small frozen particles (i.e., snow) with low fall velocities. These small frozen particles cannot fall into the warm areas and melt efficiently, resulting in a reduced melting rate. For $N_{\mathrm{CN}} < 1{,}000\ \mathrm{cm}^{-3}$, the ratio between large and small frozen particles is not sensitive to N_{CN} anymore and the vertical distribution of frozen particles become important. Increasing N_{CN} leads to earlier formation of frozen particles at low altitude, which evaporate less and result in more rainfall. We add this explanation in the main text. Please see lines 400-407.

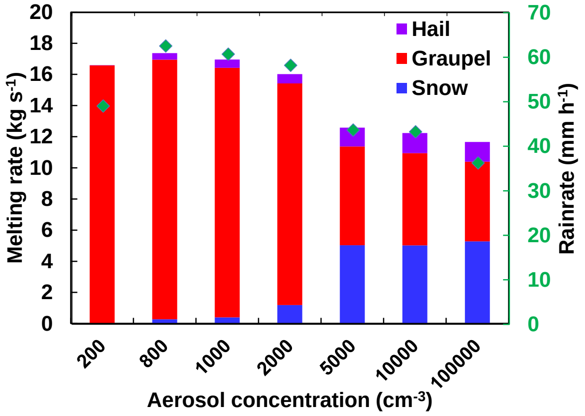


Figure R5. The correlation of rain rate and the melting rate of the frozen particles. The green diamond points are the averaged rain rate under different aerosol concentrations (*FF* = 10^5 W m^{-2}). The columns represent the integrated melting rate from individual frozen particles.

Section 3.3 I would not trust too much on those process rates since they really depend on the parameterizations of processes. I saw very different process rates between bulk and bin microphysical parameterizations and even between two 2-moment bulk schemes. Many of those sensitivities are scheme-dependent. Please discuss it.

Response: Yes, we agree with the referee. In Sect. 3.3, we add more discussion to emphasize the bias which could be caused by using different microphysical schemes. The text is “We are aware that the exact process rates may vary depending on the microphysical schemes used in the simulation (Muhlbauer et al., 2010). Therefore, we stress that the process analysis here is based on the Seifert microphysical scheme (Seifert and Beheng, 2006). In the future, further observations from laboratory and field measurements are needed to improve the understanding of aerosol-cloud interactions and to better constrain microphysical parametrizations.” “In this study, we demonstrate the performance of ensemble simulations in determining the regime dependence of aerosol effects. The use of such regime dependence requires caveats because it may differ for different cloud types, aerosol properties, meteorological conditions and model configurations (e.g., microphysical schemes, dynamic schemes, dimensionality, etc.; the 3-D results are in the supplementary material)". Please see Lines 489-494, and 739-743 respectively.

P7798, Line 5-10: cloud radiative forcing and cloud lifetime effects are not examined in this study, what do the conclusions come from? A recent study over long time scale (Fan et al., 2013) suggested significant aerosol effects on deep convective cloud morphology and lifetime.

Response: Since we did not directly calculate the forcing, here we just delete the comments on the radiative forcing and cloud lifetime. We also cite Fan et al. (2013) in this part. The text is "For this case study of pyro-convective clouds, then, we conclude that aerosol effects on cloud droplet number concentrations and cloud droplet size are likely more important than effects on precipitation, since precipitation is far less sensitive to aerosol number concentrations than to updraft velocity. This is in agreement with other studies (e.g., Seifert et al., 2012). A recent long-term convective cloud investigation found that microphysical effects driven by aerosol particles dominate the properties and morphology of deep convective clouds, rather than updraft-related dynamics (Fan et al., 2013). Therefore, it must still be determined whether this conclusion applies to other cloud types and over longer time scales." Please see lines 730-738.

Minor comments: 1. p7783 Line 16, how do you get 85 km with 110 grids of 500 meter spacing?

Response: The simulation grids are stretched, not evenly divided. Only the horizontal grid at the center of the modeling domain is equal to 500 m, and towards to the lateral boundaries the grid size becomes bigger according to the width of zoom (which was set in the input file).

2. Please use correct terminology: cloud freezing should be “drop freezing”, depositional growth of droplets should be “condensational growth of droplets”.

Response: Accepted. We have corrected these terms through the text.

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