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 #2

This manuscript investigates the impacts of cloud condensation nuclei (CCN) and dynamic condition on pyro-clouds using a 2-D Active Tracer High Resolution Atmospheric Model (ATHAM) with a double-moment cloud microphysical scheme. Wide ranges of CCN concentration and convection strength were used to configure the sensitivity simulations with totally over 1000 runs. By carefully assessing the budget and evolution of hydrometeors as well as microphysical processes rates, the paper sorted out the different sensitivity regimes for aerosol and updraft velocity individually, and potentially shed some light on physical mechanism involved in the aerosol-cloud interaction. However, there are several problems that need to be adequately addressed before the paper can be accepted for publication.

1) In the abstract, authors emphasized that aerosols suppress the surface precipitation when aerosol concentration is between 1000 to 3000 cm^{-3}. However, Li et al. (2008, JGR) showed the opposite aerosol effect during the same aerosol range for the cumulus cloud. It indicates that CCN values here are not representative as thresholds to distinguish the aerosol effect.

Response: We thank the reviewer for pointing this out. We agree that the exact threshold value is subject to certain regime and may not be universally applicable to all conditions. It may change due to the differences in the model dimensionality

(2D vs. 3D), spatial dimension (single cloud convection vs. regional cloud evolution), microphysics and etc.

We make clear of this point and include more discussion concerning these uncertainties in the main text: "The threshold to distinguish the aerosol positive and negative effects is derived from the current simulated pyro-convective clouds. The cumulus cloud investigation in Li et al. (2008) also suggested this non-monotonic trend, with the threshold aerosol value around 3000 cm^{-3}. The existence of threshold N_{CN} in both studies implies that similar cloud types may have a similar regime dependence, of which the exact shape may differ due to difference in the meteorological conditions, aerosol properties, etc." "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 389-394, and 739-743 respectively.

2) Fig. 1 doesn't deliver many messages, especially in the introduction part. I would suggest move it to the conclusion part and replace the question mark by the major findings in this study.

Response: Accepted and thanks for the suggestion. This figure helps us review the study progress of aerosol-cloud interaction at different scales and conclude our main research findings. We complete this figure and move it to the conclusion section. Please see Fig. R1, which is Fig. 23 in the revised manuscript.

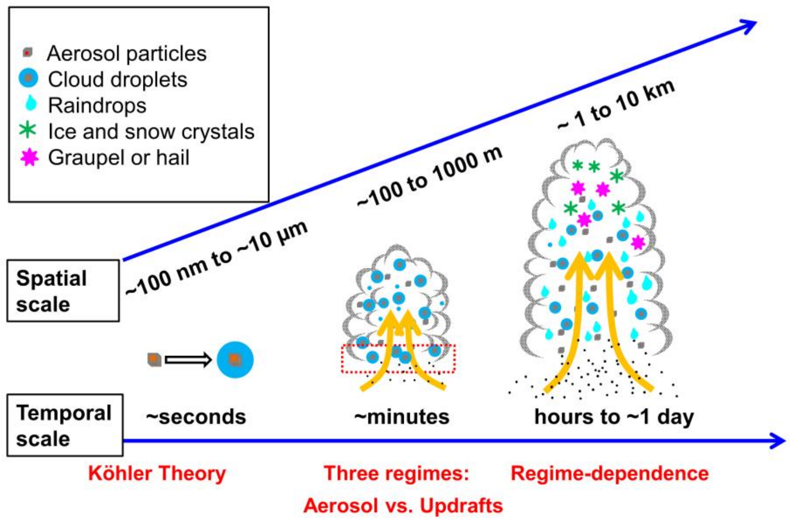

Figure R1. Overview of the research approaches on multi-scale cloud initialization and development.

3) Each simulation was conducted for only three hours. Is three-hour long enough to capture the lifetime of a typical pyro-cloud? From Fig. 12, it is clear that the precipitation was still going on after three hours.

Response: This is a good point. We agree that different time scales may change the regime dependence of aerosol effects (McComiskey and Feingold, 2012). The lifetime of deep convective clouds varies from several hours to days based on previous studies (Lindsey and Fromm, 2008; Hagos et al., 2013). Within our work, when fire forcing is weak, 3 simulation hours could cover the lifetime of most pyro-convective clouds. When fire forcing is very strong, the production of cloud hydrometeors and precipitation keeps in a steady level within 3 simulation hours (HULA, HUHA cases in Figs. 5, 9).

We have tested a longer simulation time (6 simulation hours) to examine the dependence of rain rate on aerosol concentration and fire forcing. As shown in Fig. R2, the results of 6-hour simulation are qualitatively similar to the 3-hour case (Figure 13a in new manuscript), we thus stick to the 3-hour results.

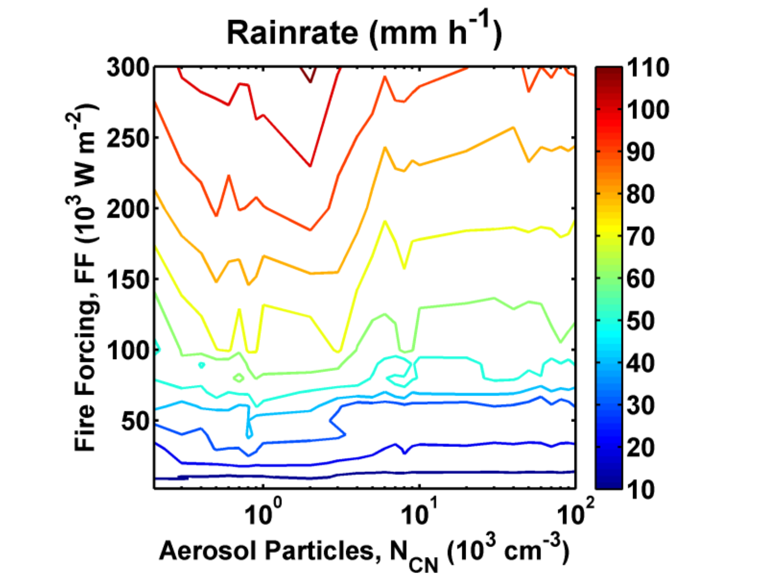

Figure R2. Contour plot of rain rate calculated as a function of aerosol number concentration (N_{CN}) and updraft velocity (represented by *FF*). Data are from the results of 6 simulation hours.

4) The prescribed aerosol budget used in this study could bias aerosol effects. Wang et al. (2013, JGR) has pointed out that prescribed aerosol scheme overestimates the magnitude of aerosol effects, and even changes the sign of aerosol effects with bulk microphysics. Similar discussion is necessary here and an implementation of a prognostic aerosol approach would be more valuable.

<u>Response:</u> Thanks for the comments. We admit that the prescribed aerosol method can lead to bias in the results. One of the main reasons is because our ATHAM model doesn't have an aerosol module. Therefore, we followed some previous studies (Seifert et al., 2012; Reutter et al., 2013) and used the prescribed aerosol distributions as an alternative. We have included more discussion about the uncertainty of prescribed aerosol approach and the importance of prognostic approach. Lines 152-156: "A similar prescribed approach has been used in previous studies (Seifert et al., 2012; Reutter et al., 2013). Some previous studies have pointed out that a prescribed aerosol scheme overestimates the magnitude of CCN concentrations compared to a prognostic aerosol scheme, because it lacks a representation of the efficient removal of particles by nucleation scavenging (Wang et al., 2013)."

5) Page 7787 line 20, the statement "As N_{CN} or FF increases, their impact becomes weaker" is not accurate. Clearly from Fig. 3b, sensitivities of cloud droplets to FF become larger after $4*10^4$ W m^{-2}.

Response: We have revised the manuscript accordingly as "High sensitivities were found for low conditions of N_{CN} and *FF*. While there are some deviations (which appear to be random numerical noise), in general, as either N_{CN} or *FF* increases, the impact on the cloud droplet number concentration of further changes to either the variable becomes weaker (Figs. 7c and 7d)." Please see lines 280-283.

6) Page 7788 line 4, the statement "when we evaluate the cloud responses to the changes in the ambient aerosol particles for global models or satellite data, we should focus more on the aerosol effect on cloud droplet number concentration, rather than on the liquid water path" is problematic. From Fig. 3c, it is clear that the sensitivities of cloud mass to CCN is quite pronounced under low updraft condition with CCN concentration less than 2000 cm^{-3}. Meanwhile, this is the typical maritime condition for stratocumulus clouds, which are prevalent over the most ocean region. Therefore, the aerosol effect on cloud liquid contend is very important.

Response: We agree and we have removed this statement in the revised manuscript.

7) Section 3.2.2, there is no physical explanation of the complicated response of the raindrop concentration to aerosols and updrafts.

Response: Sorry for the confusion. Within the main text, we put the general results in Sect. 3.2, and present the corresponding detailed physical explanation in Sect. 3.3. The microphysical explanations for raindrops will be found in Sect. 3.3.2.

8) Page 7789 line 19-22, it is reported that "greater concentrations of aerosol result in more snow and less graupel", but actually some other studies suggested that elevated aerosols could increase the graupel/hail in the convective system (Khain et al., 2009, JGR;Wang et al., 2011, ACP). This is attributed to the competing effects of aerosols on the graupel formation. Since graupel is mainly formed by the accretion of supercooled drops by ice or snow, the smaller but more abundant supercooled cloud droplets in the polluted condition could be either favorable or not for graupel formation.

Response: Thank for the comments. In the work of Khain et al. (2009) and Wang et al. (2011), more aerosols are suggested to enhance the collision between graupel particles and small supercooled droplets, and thus the graupel/hail formation. Within our work, we found only under LULA (low updrafts and low aerosol) condition, riming of cloud droplets and raindrops (*crg* and *rrg*) is an important source of graupel (Fig. R3). When aerosol concentration increases, more droplets are prone to form small frozen particles (ice and snow) firstly, and the main source of graupel is from the collection of these small frozen particles. This may explain the difference with Khain et al. (2009) and Wang et al. (2011).

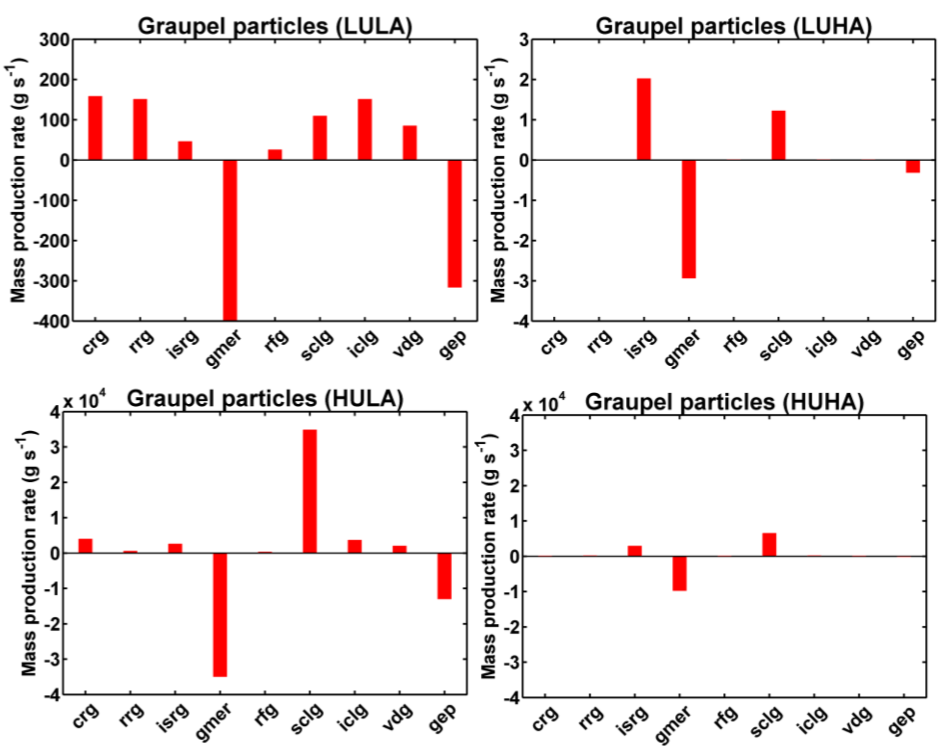

Figure R3. Comparisons of the time-averaged rates of change in graupel concentration resulting from the main processes, which were obtained from the domain-integrated values. Sources are plotted as positive values, and sinks are negative. The acronyms indicate *c/r/i/srg*: riming of cloud droplets/raindrops/ice/snow to form graupel; *gmer*: melting of graupel to form raindrops; *rfg*: freezing of raindrops to form graupel; *i/sclg*: collection of ice/snow to form graupel; *vdg*: condensational growth of graupel by water vapor; *gep*: evaporation of graupel.

In the main text, we have included more discussion to address the diverse aerosol effects on the graupel production. Please see Lines 354-359. The text is "Other research has suggested that elevated aerosols could increase the concentration of large frozen particles (graupel/hail) in the convective system (Khain et al., 2009; Wang et al., 2011), which was attributed to the competing effects of aerosols on the graupel formation. Since graupel is mainly formed by the accretion of supercooled droplets by ice or snow, the smaller but more abundant supercooled drops under polluted conditions could be either favorable or unfavorable for graupel formation."

9) It is nice to see that authors stress the importance of a longer period simulation. Actually, Fan et al. (2013, PNAS) and Wang et al. (2014, Nature Communication) have done some long-term (more than one month) cloud-resolving modeling studies over certain cloud regions. Please discuss accordingly.

Response: Fan et al. (2013) and Wang et al. (2014) suggested that anthropogenic aerosols will increase accumulated rain trend. They also conclude that the most important influence induced by aerosols is the redistribution of precipitation, indicated by the reduced light rain occurrence frequency and increased heavy rain frequency in polluted regions. This could lead to higher risk of droughts and floods in monsoon regions due to more serious pollution. In the revised manuscript, we add the following discussion on lines 416-418: "Simulations for a longer period should be carried out in future studies to investigate the influence of aerosols on precipitation over longer time scales as in Fan et al. (2013) and Wang et al. (2014)."

10) I'm concerned about the way authors calculate the microphysical process rates. Since the rates are averaged over the whole domain, I would expect that the cloud occurrence/fraction over the domain might significantly affect the microphysical rates there. It will be important to report the rates from cloud-only-points as well.

Response: We have been thinking of both ways of calculating process rates. The reason that we finally made the average over the domain is to be consistent with the way we calculated the averaged number/mass concentration. Otherwise, the rate and concentration will not be directly comparable due to the influence of cloud occurrence/fraction.

11) In Fig. 13 and 15, g/h/s/imer should be melting to form raindrops, rather than "multiplication to form ice crystals".

Response: Accepted. We corrected this explanation. Please see Figs. 18 and 20 in the revised manuscript.

12) In Fig. 11 and 13, it shows that autoconvertion rate from cloud droplets to raindrops is higher in the high aerosol scenario (HA) than that in the clean case (LA). Why?

Response: The domain-averaged rate is in fact slightly reduced as N_{CN} increases (LA→HA) under high updraft (HU) condition. The apparent difference may result from different scales used in these figures.

13) Page 7795 line 7, what is the reason behind the phenomena "although snow is the dominant constituent of frozen particle mass (Fig. S4), the deposition of vapor on ice (vdi) rather than on snow is the major pathway for frozen particles"?

Response: After examining the budge of snow, the process of collecting of ice to form snow (processes of *iscs*, and *icls*) is much more efficient than other source processes (Fig. R4), which are internal conversions not counted as either a source or a sink of frozen water content. The ice crystals used for conversion to snow is mostly from the deposition of vapor on ice (*vdi*). Once small ice crystals appear, they can quickly collide to be snow. We add this explanation in the main text, and please see Lines 571-574: "The increase of snow mass is mostly caused by collecting of ice (*ics*) and ice self-collection (coagulation of ice particles, *iscs*), which are internal conversions not counted as either a source or a sink of frozen water content. The ice crystals used for conversion to snow derive mostly from the *vdi* process".

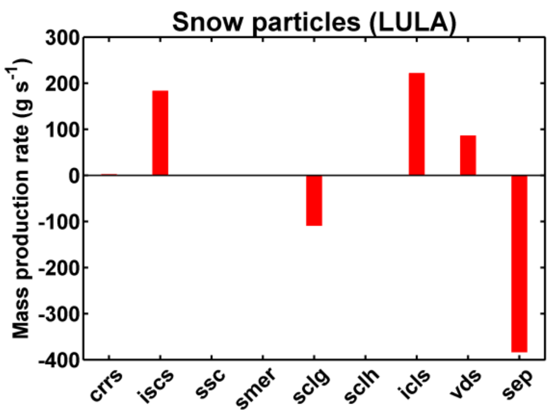

Figure R4. Comparisons of the time-averaged rates of change in snow concentration resulting from the main processes, which were obtained from the domain-integrated values. Sources are plotted as positive values, and sinks are negative. The acronyms indicate crrs: riming of cloud droplets and raindrops to form snow; iscs: selfcollection of ice to form snow; ssc: selfcollection of snow; smer: melting of snow to form raindrops; sclg/h: collection of snow to form graupel/hail; icls: collection of ice to form snow; vds: condensational growth of snow by water vapor; sep: evaporation of snow.

References

Fan, J. W., Leung, L. R., Rosenfeld, D., Chen, Q., Li, Z. Q., Zhang, J. Q., and Yan, H. R.: Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds, P Natl Acad Sci USA, 110, E4581-E4590, doi: 10.1073/pnas.1316830110, 2013.

Hagos, S., Feng, Z., McFarlane, S., and Leung, L. R.: Environment and the Lifetime of Tropical Deep Convection in a Cloud-Permitting Regional Model Simulation, J Atmos Sci, 70, 2409-2425, doi: 10.1175/Jas-D-12-0260.1, 2013.

Khain, A. P., Leung, L. R., Lynn, B., and Ghan, S.: Effects of aerosols on the dynamics and microphysics of squall lines simulated by spectral bin and bulk parameterization schemes, J Geophys Res-Atmos, 114, doi: 10.1029/2009jd011902, 2009.

Li, G. H., Wang, Y., and Zhang, R. Y.: Implementation of a two-moment bulk microphysics scheme to the WRF model to investigate aerosol-cloud interaction, J Geophys Res-Atmos, 113, doi: 10.1029/2007jd009361, 2008.

Lindsey, D. T., and Fromm, M.: Evidence of the cloud lifetime effect from wildfire-induced thunderstorms, Geophys Res Lett, 35, doi: 10.1029/2008gl035680, 2008.

McComiskey, A., and Feingold, G.: The scale problem in quantifying aerosol indirect effects, Atmospheric Chemistry and Physics, 12, 1031-1049, doi: 10.5194/acp-12-1031-2012, 2012.

Reutter, P., Trentmann, J., Seifert, A., Neis, P., Su, H., Herzog, M., Wernli, H., Andreae, M. O., and Poschl, U.: 3-D model simulations of dynamical and microphysical interactions in pyro-convective clouds under idealized conditions, Atmos Chem Phys Disscuss, 13, 19527-19557, doi:10.5194/acpd-13-19527-2013, 2013.

Seifert, A., Kohler, C., and Beheng, K. D.: Aerosol-cloud-precipitation effects over Germany as simulated by a convective-scale numerical weather prediction model, Atmospheric Chemistry and Physics, 12, 709-725, doi: 10.5194/acp-12-709-2012, 2012.

Wang, M., Ghan, S., Ovchinnikov, M., Liu, X., Easter, R., Kassianov, E., Qian, Y., and Morrison, H.: Aerosol indirect effects in a multi-scale aerosol-climate model PNNL-

MMF, Atmospheric Chemistry and Physics, 11, 5431-5455, doi: 10.5194/acp-11-5431-2011, 2011.

Wang, Y., Fan, J. W., Zhang, R. Y., Leung, L. R., and Franklin, C.: Improving bulk microphysics parameterizations in simulations of aerosol effects, J Geophys Res-Atmos, 118, 5361-5379, doi: 10.1002/Jgrd.50432, 2013.

Wang, Y., Zhang, R. Y., and Saravanan, R.: Asian pollution climatically modulates mid-latitude cyclones following hierarchical modelling and observational analysis, Nat Commun, 5, doi: 10.1038/Ncomms4098, 2014.