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".

Response to Anonymous Referee #4

This study used a 2D atmospheric model with a 2-moment microphysical scheme to simulate pyro-clouds. The effects of aerosol and convection intensity on cloud, rain, ice-phase particles, as well as surface rainfall were studied using a test matrix of 31 aerosol concentrations by 42 convection intensities. The authors also carried out process analysis for 4 individual simulations to explore mechanisms of the simulated sensitivities. Results from these process analyses essentially agreed with various previous studies, although nothing new was found. The strength of this study, in my opinion, is the large number of sensitivity simulations which afford more robust sensitivity analyses. However, the authors did not take the full advantage of their simulations. For example, they reverted back to analyzing only 4 individual members in their PA analysis, instead of studying the mean and variations of all available members. Since there is a large room for improvement here, I would recommend publication with major revision. I hope the authors will take full advantage of their large simulation dataset and add more depth to their analyses.

<u>Response:</u> Thanks for the constructive suggestions. In the revised manuscript, we extend the process analysis from four individual cases to the full interested ranges of aerosols and fire forcing in a way as shown in Fig. R1 and Figs. 19, 21, 23 in the revised manuscript.
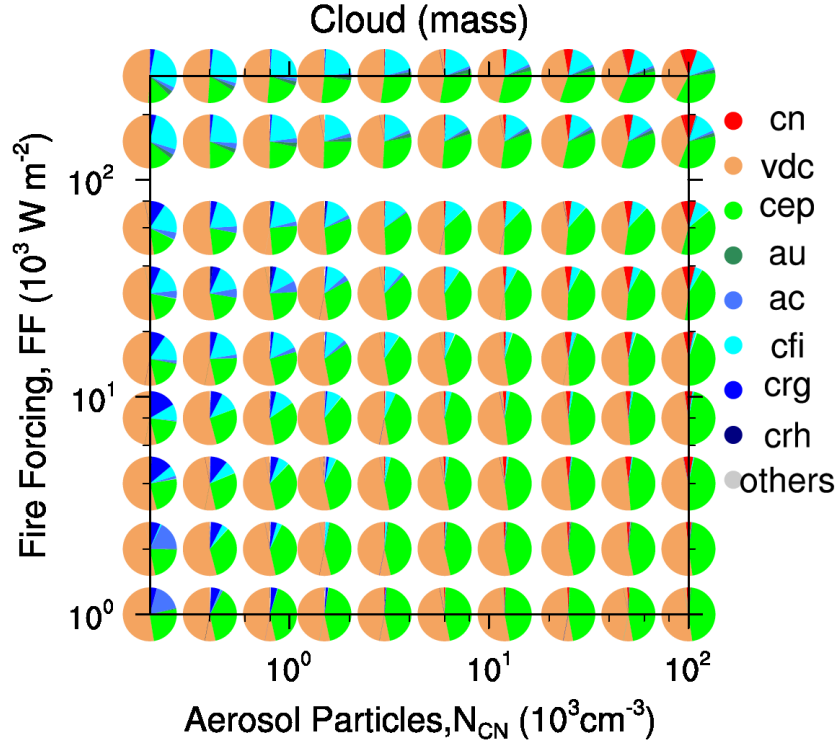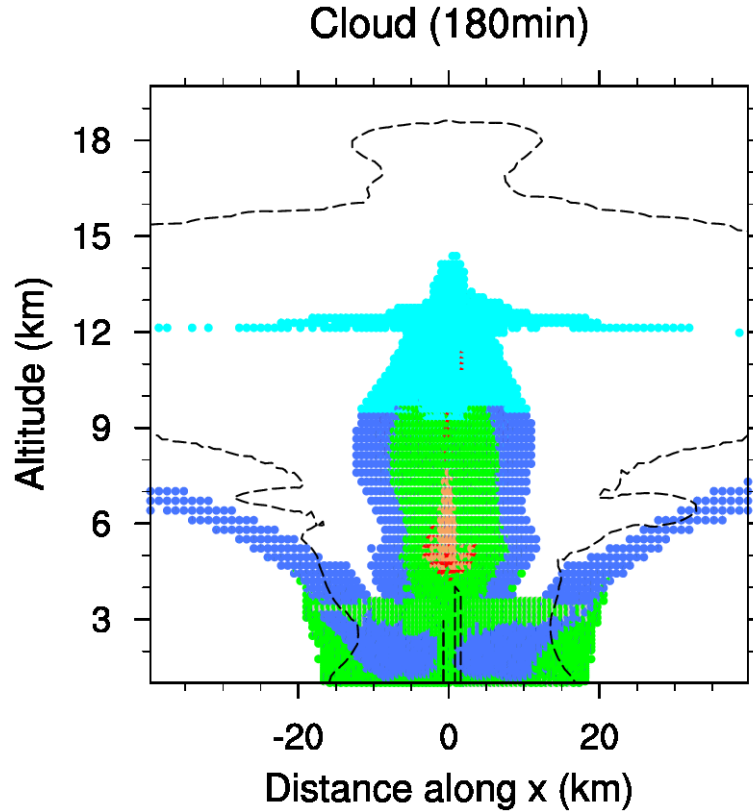
Figure R1. The pie charts summarize the relative percentage of the microphysical processes involving cloud droplets as a function of N_{CN} and fire forcing. Colors within each pie chart reflect the contribution of processes under the specific condition. Warm colors denote the source, while cold colors 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.

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. This provides a whole picture that how the contribution of each processes evolve as aerosol concentration or fire forcing increases.

Besides, we also plot the vertical cross sections of the change rate of these microphysical processes contributing to cloud water content in the modeling domain and the temporal evolution of the contributions (e.g., Fig. R2). The

figures for all the simulation period are in Figs. 20, 22 and 24 for cloud drop-
lets, raindrops and frozen particles respectively. The corresponding analysis
has been included in Sect. 3.3.

Cloud (180min)

Figure R2. The pie charts summarize the vertical cross sections of the change rate of main microphysical processes contributing to cloud water content. Each pie chart shows the averaged contribution over the past 30 min. Colors within each pie chart reflect the percentage of processes in each grid. The black dashed line is the 0.1 μg kg^{-1} isoline of the interstitial aerosol, indicating the shape of smoke plume. The meaning of the acronyms is the same as in Fig. R1.

Major concerns:

1. Convection is a highly non-linear process. This puts a serious constraint on individual sensitivity studies. One of the ongoing debates is how representative such an individual case study is in elucidating aerosol-cloud interactions. With >1000 simulations and independent variations in two external forcings (aerosol and fire intensity), this study may

be able to shed some light on these debates. For example, if we were to conduct 2 sensitivity tests with high/low CN number (e.g., 2x or10x aerosol concentration), what is the probability that we will be able to get RS within one standard deviation from the mean? Will we be able to at least get the RS sign correctly? How robust is it to apply the mechanisms derived form an individual case with contrasting aerosol scenes to various environmental conditions, in this case, fire intensity? Statistical analysis along this direction will be very helpful in quantifying uncertainties of individual studies. It could also guide designs of future sensitivity tests.

<u>Response:</u> I appreciate the comment very much. As mentioned in the comment, aerosol-cloud interactions are regarded as nonlinear processes. In this case, the local aerosol effects on a cloud relevant parameter Y, i.e., dY/dN_{CN} can be different from $\Delta Y/\Delta N_{CN}$, the dependence derived from two case studies. In Sect. 3.4 of the revised manuscript, we try to answer how much difference can be expected between dY/dN_{CN} and $\Delta Y/\Delta N_{CN}$. In the following, we take the responses of the precipitation to aerosols for example to address this issue.

Figure R3 (Fig. 22 in revised manuscript) shows the statistics of the relative difference between $\Delta Y/\Delta N_{CN}$ and dY/dN_{CN} under LU and HU conditions, in which *Y* represents the precipitation rate. As precipitation is insensitive to aerosols for $N_{CN} > 10,000$ cm^{-3}, only the cases with N_{CN} of 200~10,000 cm^{-3} are chosen in the calculation. The relative difference is defined as:

$$\text{Relative difference} = \frac{\dfrac{\Delta Y}{\Delta N_{CN}} - \dfrac{dY}{dN_{CN}}}{\dfrac{dY}{dN_{CN}}}$$

and $\dfrac{\Delta Y}{\Delta N_{CN}}$ is calculated as: $\dfrac{\Delta Y}{\Delta N_{CN}} = \dfrac{Y(2N_{CN}) - Y(N_{CN})}{2N_{CN} - N_{CN}}$, in which the aerosol effect is determined by the difference between the reference case and that after doubling N_{CN}. $\dfrac{dY}{dN_{CN}}$ is the derivative of the precipitation rate at each N_{CN}, representing the local dependence of precipitation on N_{CN}.

The histograms in Fig. R3 demonstrate that $\dfrac{\Delta Y}{\Delta N_{CN}}$ can deviate considerably

from $\dfrac{dY}{dN_{CN}}$, not only for the absolute value but also for the sign. Statistically, most of the relative differences are in the range of -3.7~0.9 (the 25th and 75th percentiles respectively, with the average difference of -3.0) under LU condition, while are between -1.5 and 0.04 (the 25th and 75th percentiles respectively, with the mean value of 0.02) under HU condition. The fact that individual case studies may not reveal local aerosol effects demonstrates the importance of ensemble studies in determining the real responses of clouds to aerosol perturbations.
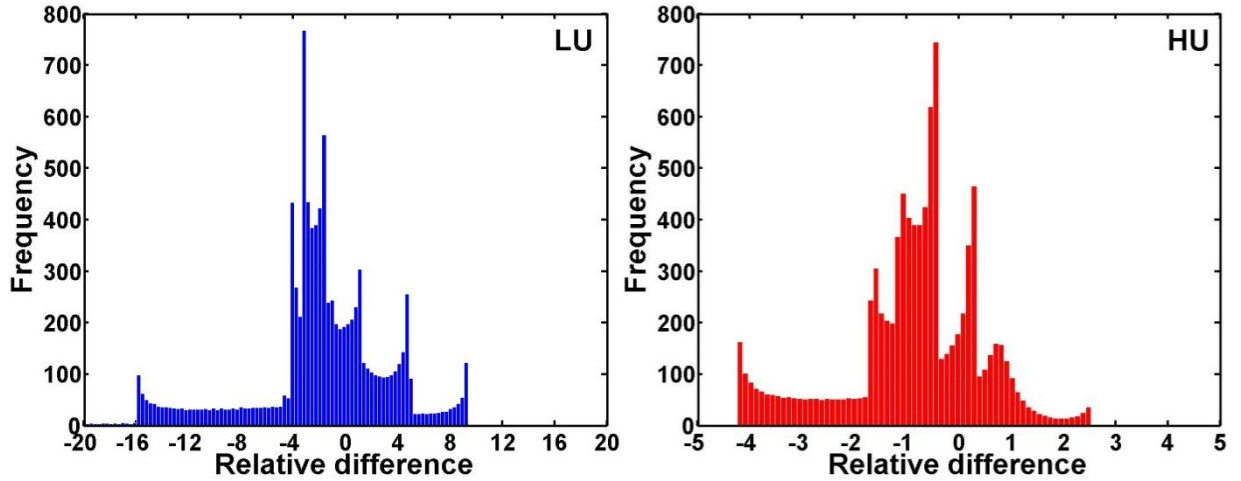

Figure R3. Histograms of the relative difference between $\dfrac{\Delta Y}{\Delta N_{CN}}$ and

$\dfrac{dY}{dN_{CN}}$ under LU and HU conditions, where Y here denotes precipitation

rate. $\dfrac{\Delta Y}{\Delta N_{CN}} = \dfrac{Y(2N_{CN}) - Y(N_{CN})}{2N_{CN} - N_{CN}}$, and $\dfrac{dY}{dN_{CN}}$ is the derivative of the pre-

cipitation rate along the variable N_{CN}.

For other hydrometeors, we also get such relative difference figures following this method, and found individual case studies are largely biased from the local derivatives. Different selection of the parameter space may result in different or even opposite conclusions. Therefore, our continuous sensitivity study over a wide range of parameter space shed some lights on these debates. Concerning the length of the manuscript, we just include the discussions for rain rate in the revised manuscript. Please see section 3.4.

2. There is an inconsistency in the RS analysis in the first part, which used 1302 cases, and the PA analysis in the second part, which used only 4 individual simulations. How do we know that mechanisms derived from PA analysis for an individual case are the same mechanisms that produced the mean sensitivities for hundreds of cases? If the authors can prove that the 4 individual cases are representative (see my comments in the previous paragraph), future aerosol-cloud simulations may be greatly simplified. If this cannot be proven, then PA analysis need to be done the same way as RS analysis, using all 1302 simulations.

Response: In the revised manuscript, we extend the process analysis from four individual cases to the full interested ranges of aerosols and fire forcing. The percentage of the microphysical processes under different aerosol and fire forcing conditions would be presented in the revised manuscript. Please see Fig. 19, 21, 23, and the text is in Sect. 3.3. Take cloud droplets for example, the figure is like this:
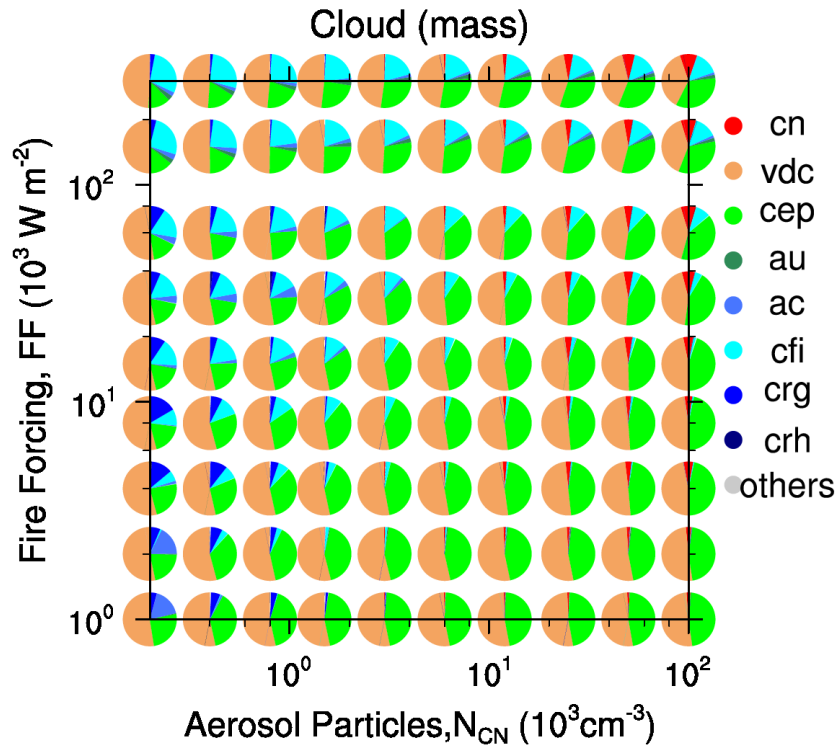
Figure R4. The pie charts summarize the relative percentage of the microphysical processes involving cloud droplets as a function of N_{CN} and fire forcing. Colors within each pie chart reflect the contribution of processes under the specific condition. The acronyms indicate cn: cloud nucleation; vdc: 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.

Minor points:

1. The current simulation used pyro-cloud set up, e.g., there is a steady heat source at surface. This is fundamentally different from, e.g., a cumulus formed in the atmosphere. The authors should limit their discussions within pyro-clouds. Certain speculative comments, e.g., P7788, L2, P7798, L1, may not be applicable. I would suggest removing them from the discussion.

Response: Thanks for the comments. We have removed these sentences "This strongly suggests that when we evaluate the cloud responses to the changes in the am-

bient 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.", and revised the sentence "For this case study, then, we conclude that aerosol effects on cloud droplet number concentrations and thus cloud radiative properties (first indirect effect) are likely more important than effects on precipitation and thus cloud lifetime (second indirect effect), since precipitation is far less sensitive to aerosol number concentrations than to updraft velocity." to be "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 N_{CN} than to updraft velocity." Please see Lines 730-733.

In addition, we have also investigated how the cloud and precipitation evolve if the fire forcing was shut down after half hour. The contours for each hydrometeor and precipitation are shown in Figs. R5, R6, R7, and R8 (not shown in the revised main text). For the domain-integrated concentration, the dependences of individual hydrometeor on aerosol concentration and fire forcing ended up showing good agreement with the simulations with persistent fire forcing. We included this information in the revised manuscript: "These results are derived from the simulations with persistent fire forcing over modeling period. We have also examined the case in which the fire forcing was shut down after the first half hour of simulation (not shown). The same regimes were found in these simulations, with boundaries in good agreement with the findings presented in this work." Please see Lines 296-300.
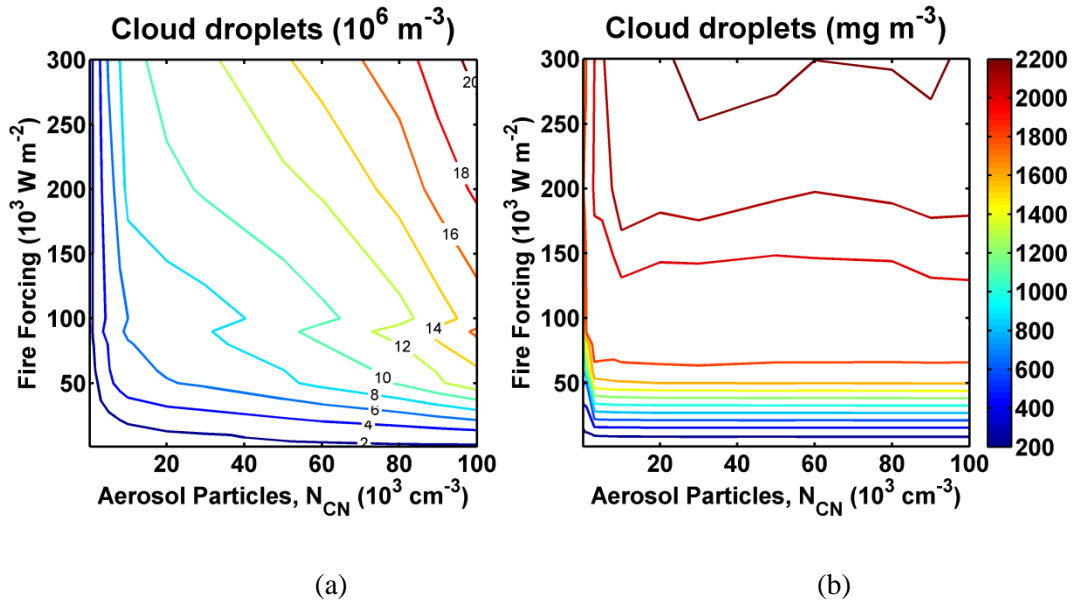
Figure R5. Number (a) and mass concentration (b) of cloud droplets calculated as a function of aerosol number concentration (N_{CN}) and updraft velocity (represented by FF).
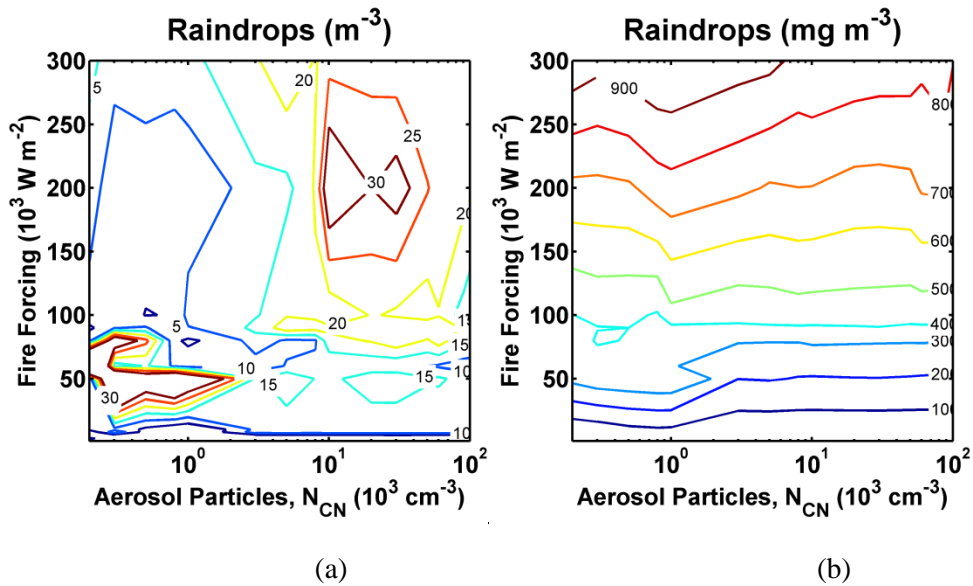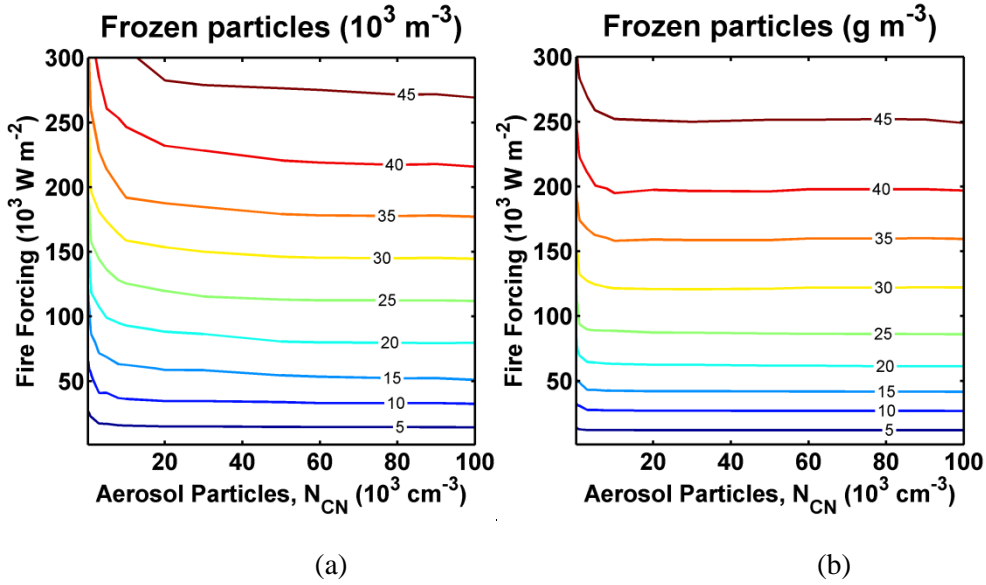

Figure R6. Same as Fig. R5 but for raindrops.

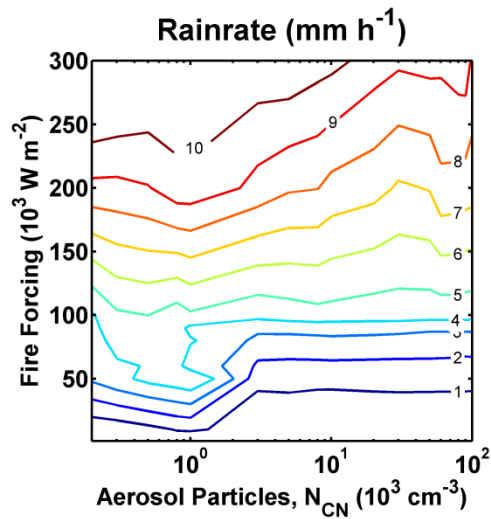**Figure R7. Same as Fig. R5 but for frozen particles.**

Figure R8. Same as Fig. R5 but for rain rate.

2. In the sensitivity tests, CN concentration ranges from 200 to 100000 per cubic centimeter, fire intensity ranges from 1000 to 100000 W/m^2. Can you describe what ranges of CN and fire intensity are realistic? Does higher fire intensity also produce higher CN? Obviously 200 cm^-3 is not realistic in any pyro clouds. This can guide the readers to pay

more attention to certain ranges of the parameter space. This information should be added explicitly in section 2.2.

Response: Yes, the condition with low aerosol and weak updrafts is not representative for a real pyro-convective cloud, and is used here for sensitivity studies. More CN will be emitted as fire forcing goes up. We have included the following sentence to avoid misleading the readers: "In reality, the composition and quantity of biomass burning emissions depend on the moisture content of fuels, combustion conditions, weather situation, and fire behavior (Bytnerowicz et al., 2009). What's more, the biomass burning plumes can in turn change the relative humidity as well. The aerosol particle number concentrations in biomass burning plumes usually exceed 10^4 cm^{-3}, and can be up to $\sim 10^5$ cm^{-3} (Andreae et al., 2004; Reid et al., 2005). In contrast to regular convection, the updraft velocities in pyro-convective clouds are normally larger than 20~30 m s^{-1} (Khain et al., 2005). On the basis of these facts, within our work more attention is paid to situations with higher aerosol concentration ($>10^4$ cm^{-3}) and strong updrafts (>20 m s^{-1}), which are more representative of pyro-convective clouds." Please see Lines 170-179.

3. RS values show large fluctuations for fire forcing between 2x10ˆ4 to 1x10ˆ5 (fig. 3,5,7,9). The authors could do more study on why this is the case. For example, do these fluctuations occur during the initial formation of the pyro clouds? Since the model used a steady heating at the surface, using results from the last hour may reduce these fluctuations.

Response: Thanks for the comments. We investigated the temporal evolution of the cloud hydrometeor, and found the fluctuations between 2×10^4 to 1×10^5 W m^{-2} are due to the occurrences of secondary cloud during the simulation period. Take the sensitivity of cloud water content to fire forcing for example, we picked up 4 points along the sensitivity line for HA case to check how the concentration varies. These four points are marked in Fig. R9 by green mark-

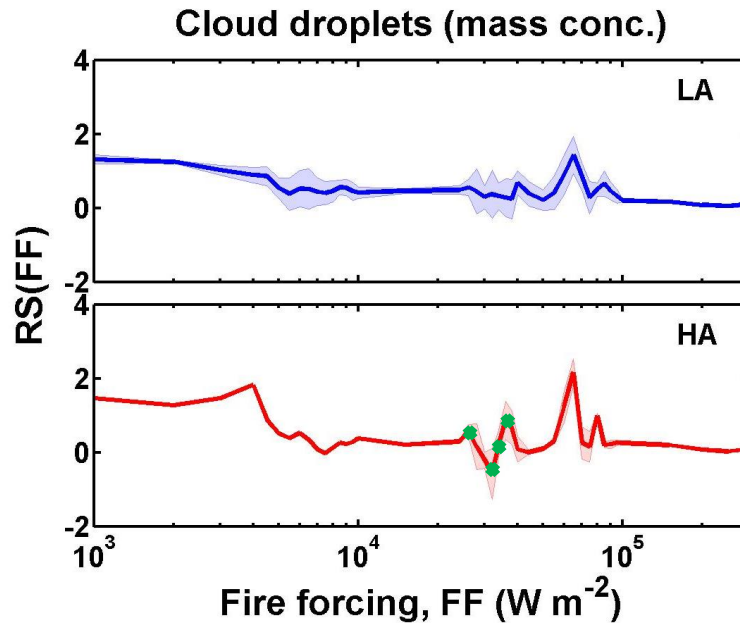ers, which correspond to *FF*= 26,000, 32,000, 34,000 and 36,000 W m^{-2} respectively.

Figure R9. Relative sensitivities with respect to *FF* for mass concentration of cloud droplets under different conditions. The thick solid lines represent the mean values under a given condition, and the shaded areas represent the variability of estimation (±½σ). The acronyms indicate LA: low aerosols (200–1,500 cm^{-3}); HA: high aerosols (10,000–100,000 cm^{-3}).

The temporal evolutions of cloud droplets for these four points are in Fig. 10. It shows that the large fluctuation is caused by the cycling of cloud formation. The simulation covers the whole period of the first cycle but only part of the second cycle. Since we are not integrating the whole cloud period, more fluctuation is introduced.
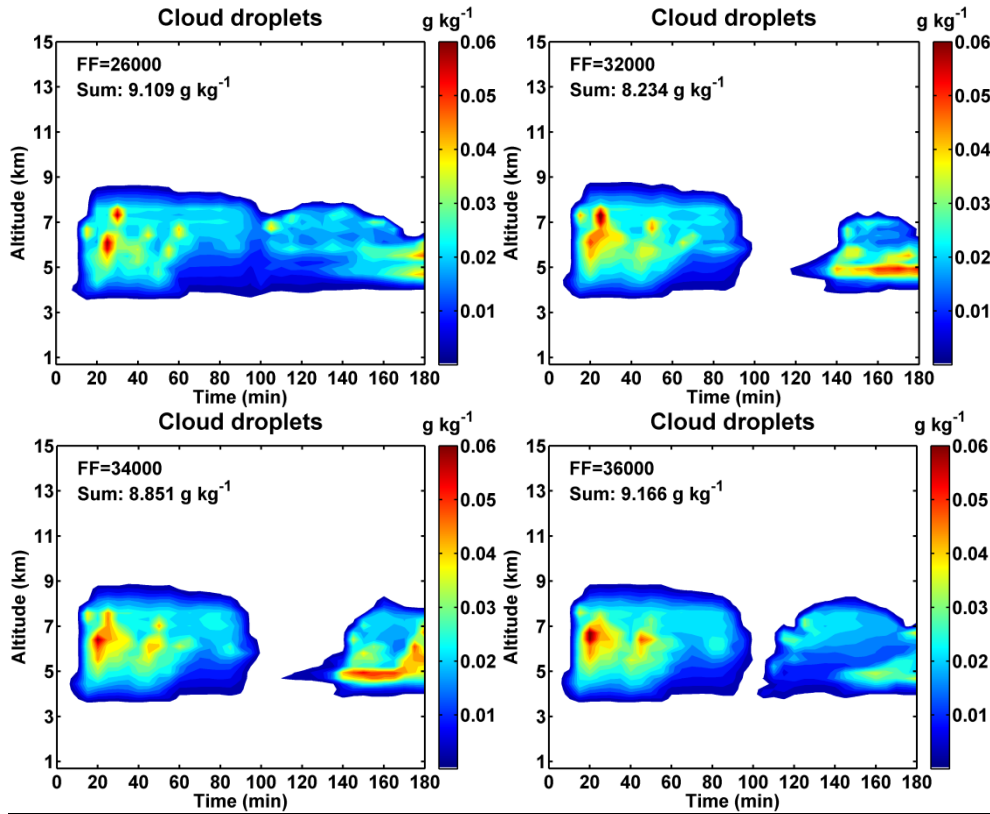
Figure R10. Time evolution of horizontally-averaged cloud water content (g kg^{-1}) as a function of altitude under different fire forcing (*FF*) conditions.

Since it appears that the first cloud usually end around 100 minutes, we plot the sensitivity of each hydrometeor to the fire forcing using the results over 0~100 min (Fig. R11). It is found, compared to the original figures, the sensitivities gets smoother for cloud droplets, raindrops, and frozen particles.
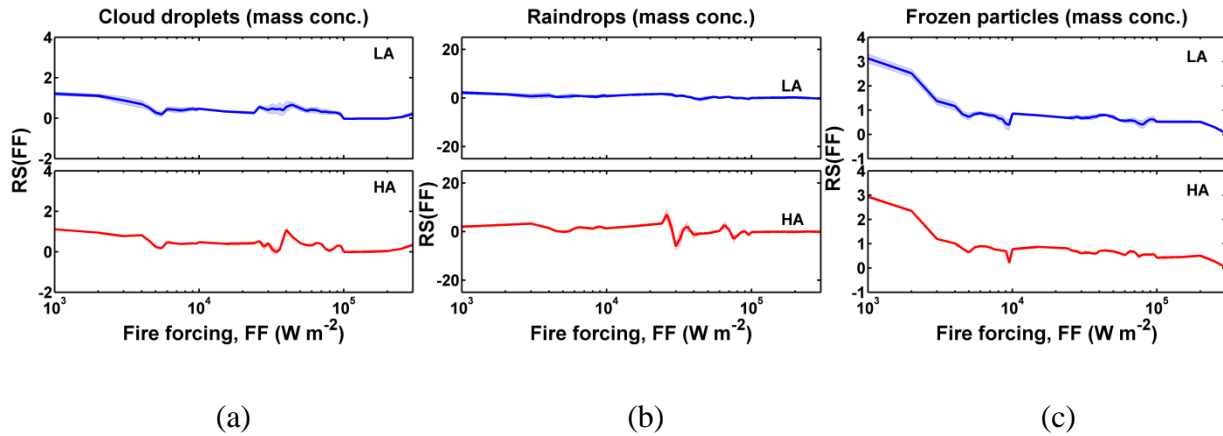
Figure R11. Relative sensitivities with respect to *FF* for mass concentration of cloud droplets (a), raindrops (b), and frozen particles (c) under different conditions. The thick solid lines represent the mean values under a given condition, and the shaded areas represent the variability of estimation ($\pm\frac{1}{2}\sigma$). The acronyms indicate LA: low aerosols (200–1,500 cm^{-3}); HA: high aerosols (10,000–100,000 cm^{-3}).

But for the precipitation rate, there remain large fluctuations (Fig. R12). This is probably because that the precipitation usually takes place at the very late period, and needs longer time. The first peak has not completed during 0~100 min.
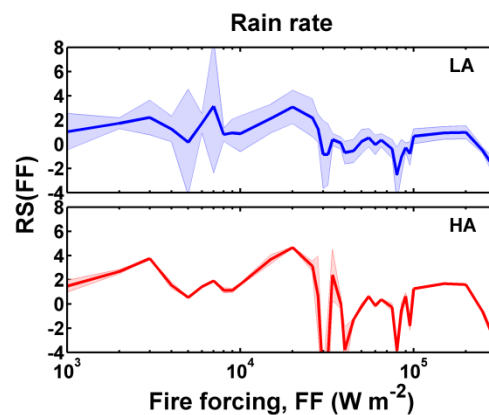

Figure R12. Relative sensitivities with respect to *FF* for rain rate under different conditions.

So far we haven't found a better way of sampling (we also tried the last hour) and thus stick to the original method.

4. P7783, L27, Is the fire forcing at a single point? What are the justifications for using a single point heating? Intuitively I thought forest fires spread to a large area, certainly larger than the 85 km domain size.

Response: The case of pyro-convection modeled in this study is based on the Chisholm

forest fire (Luderer, 2007), which is well-documented. The fire front was ap-
proximately linear, and extended from south-south-east to north-north-west.
The length of fire front is about 25 km, and the width of the fire front was
about 500 m. The 2-D simulations within our work were performed at the
cross section of the fire front, and thus only the width of the fire front was
considered (x axis). Therefore out simulation domain (85 $\times$ 26 km in the x and
z directions) can cover the fire area.

*5. P7780, L12: "When we upscale the activation of a single aerosol…", "extend" should
probably replace "upscale".*

Response: Accepted.

*6. Fig. 11, 13, 15: The scales of y-axis are all different. The authors should point that out
explicitly, instead of just tucking them discretely at the corner of each plot. If the authors
decided to calculate averages instead of 4 contrasting simulations, as I suggested in my
major concern, the mean values might be closer to each other. And the y-axis might be
more uniform for labeling.*

Response: Thanks for the suggestions. In the revised manuscript, we extend the process

analysis from four individual cases to the full interested ranges of aerosols and
fire forcing. We plotted the percentage of the microphysical processes under
different aerosol and fire forcing conditions in the revised manuscript. Please
see Fig. 19, 21, 23, and the text is in Sect. 3.3. The figure was shown at the
beginning of this response.

References

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