Interactive comment on "An explicit study of aerosol mass conversion and its parameterization in warm rain formation of cumulus clouds" by Sun et al.

We would like to thank reviewers' fruitful comments which accounted for the improvement of the quality of this work. We have thoroughly considered all of them, and have modified the manuscript accordingly. For convenience, we have put reviewers' comments in italic and our replies in regular fonts.

Anonymous Referee #2

This paper determines computationally efficient parameterizations for in-cloud aerosol processing due to collision-coalescence and accretion. The parameterizations are based on the numerical results of explicit modeling of the microphysical process rates and subsequent linear regression analysis. The regression analysis provides a set of power-law relationships (because regressions were performed in relation to the natural logarithm of the chosen variables). In practice, such relations are useful in advanced numerical modeling studies with large computational domains or over long time periods. However, there are several important shortcomings with the current study that must be addressed.

Reply: We appreciate these comments. We are very thankful for your fair comments. We have revised the manuscript according to your suggestions. We hope that the revised manuscript can be suitable for publication.

Major Comment

1. *Power-law relationships are common in the field of atmospheric science. However, they are often only useful if a general power-law formulation can be determined. This study provides a set of 64 regression equations representing two processes. Moreover, it is not particularly clear how the authors intend the equations to be used. A quick glance at the set of regression equations is enough evidence that the equations are not general and thus one must determine which equation to use for any particular situation. In other words, these equations are not useful for large domain or long time-scale modeling of aerosol-cloud interactions. The authors need to provide a way to implement the parameterization(s) or provide a much smaller set of parameterized regression equations for the reader to utilize.*

Reply: We appreciate these comments. In the bulk microphysics, the autoconversion rate and the accretion rate of cloud droplets are normally applied to calculate the precipitation production rate. For consistency with those in the aerosol-cloud interaction simulations, it is necessary to find corresponding rates for aerosol conversions. We gave many power-law equations in our old manuscript in order to analysis the possible errors due to the methodology of the parameterizations for the precipitation production rates. In the new manuscript, the regression equations for the total aerosol conversion rate with the complete coalescence approach (CCA) and the partial coalescence approach (PCA) had been deleted since those parameterizations can be approximately or exactly determined by the combination of the parameterized conversion rates for both autoconversion and accretion. Therefore, considering different concentrations and size distributions of aerosols, we gave 14 power-law formulations for the autoconversion process and accretion process, respectively, which are applicable for any aerosol-cloud interaction studies because the typical aerosol size distributions have already been considered in our parameterizations. Furthermore, according to our study, the aerosol mass conversion rates in the warm rain formation mainly depend on the aerosol concentration and the aerosol distribution shape. Different aerosol size distributions must result in the different power-law relationships for aerosol conversions. Therefore, it is difficult to find one general relationship between aerosol conversion rates and precipitation production rates to approximately handle with the aerosol conversions. So we suggested that it should be better to parameterize aerosol conversions for some typical aerosol size distributions individually rather than a general one. Depending on the aerosol size distributions in different places, we can choose suitable parameterized power-law formations for large domain or long time-scale studies.

Furthermore, the power-law relations are dependent upon ratios between a given process and a bulk aerosol or cloud property. By using these ratios in the regression equations, the authors assume that the given process and bulk aerosol/cloud property ought to have the same power except with an opposite sign. What is the rationale for this assumption? Moreover, would it not make more sense to separate these terms and perform the regression for more than one variable? In fact, several variables ought to influence the aerosol processing rates within a cloud.

Reply: We appreciate these comments. It is the most constructive suggestion to consider other variables for aerosol conversion parameterizations. As shown in the old manuscript, for the accretion process, the ratios between the intensity of the aerosol mass accretion rate and the intensity of the cloud water mass accretion rate are close to 1.0, and the same power for the cloud mass accretion rate and the cloud mass concentration can create a nice power-law regression, which means that the aerosol mass accretion rate is nearly linearly proportional to the aerosol mass concentration in cloud droplets and also proportional to the ratio of the cloud water mass accretion rate to the cloud mass concentration. However, for the autoconversion process, the aerosol mass autoconversion rate is not only dependent on those described in the old manuscript but also highly influenced by the cloud water mass accretion rate. Therefore, in the new manuscript, the aerosol mass autoconversion rate has been parameterized as a power-law function of the aerosol mass concentration in cloud droplets, the cloud water autoconversion rate, the cloud water mass concentration and the cloud water accretion rate. The new parameterized power-law equations are much better than previous schemes.

2. *Other than the scatter plots in Figures 7, 9, 11, and 16, there is very little evidence that the parameterizations actually work. In fact, some of these scatter plots would suggest that the parameterization(s) may induce large errors in the aerosol processing rates in clouds (see Figure 9d). Somewhat related to the first point above, even though a regression equation works for one setup, it may not work for another initialization. Additional simulations to verify the generality of the resulting regression equations should be performed. Regardless, simulations demonstrating the applicability of a parameterization are necessary. For example, the parameterization(s) could be implemented into a numerical model (e.g., CRM or LES) and used to determine their ability to reflect either more detailed aerosol processing calculations or observations.*

Reply: We appreciate these comments. As mentioned in our manuscript, The accuracy of simulations for the aerosol in-cloud scavenging are not only dependent on the aerosol mass conversion parameterizations but also highly related to the schemes of the precipitation production rates. In order to verify the generality of our regression equations, we are modifying our column model from the bin microphysics into the bulk microphysics with some current schemes of the precipitation production rates to minimize the difference from the simulations with the bin microphysics. Furthermore, the new aerosol conversion parameterizations and the selected precipitation production rates will be implemented into the bulk microphysics of WRF for the large-eddy simulations and also will be compared with the in-situ observations. These results will be submitted on another paper.

3. *The model used in this study may not be sufficient for determining the power-law relations. In particular, the use of Bott (2000), in which the aerosol processing is condensed into a single dimensional problem and then relaxed back to two dimensions, may induce errors in the calculation of aerosol processing.*

Reply: We appreciate these comments. In our new manuscript, the 2-D spectrum scheme of Lebo (2011) had been implemented into our model instead of Bott (2000).

Furthermore, an important part of cloud-aerosol interactions is the activation process. There are numerous methods in the literature for determining both the number and mass of activated aerosols. However, this is never discussed in the manuscript.

Reply: As we know in the real atmosphere, with air humidity increasing, the hygroscopic aerosols, such as cloud condensation nuclei, will deliquesce to solution drops, which may proceed to grow toward their critical radius for activation and subsequent quick growth. Therefore, the activation of aerosols has been normally determined with the number and mass of activated aerosols for cloud droplet calculation (Sun and Ariya, 2006). However, in some cloud models with high-resolution bin microphysics (Wobrock et al., 1998; Leroy et al., 2006) as well as our model in which the microphysics are described in detail by Sun et al. (2012), the nucleation of water droplets is represented in a continuous transition from haze droplets through diffusion growth as the way in the real atmosphere, which allows the evolution of the individual aerosol particles to followed through the water droplet spectrum as it considers only one number density distribution function, f(m_water, m_aerosol) , for all aerosol particles and water droplets together. The information on the aerosol nucleus and the attached water mass are followed via two coordinates: m_water the water droplet mass, and m_aerosol the mass of the dry aerosol nucleus. Such a treatment for hygroscopic growth of aerosols requires high-resolution bins and corresponding small time steps for numerical simulations.

The authors provide a section on the sensitivity to the threshold radius for collision-coalescence. I would suggest that the authors use a collision-coalescence parameterization that does not rely on a threshold parameter, especially in light of the large sensitivity to the chosen threshold in the manuscript.

Reply: We appreciate these comments. We modified our manuscript as your suggestions.

4. *Much of the discussion in the text revolves around the E1 case. In this case, there are approximately 1300 cm−3 aerosols. Without sufficiently large updrafts and under normal circumstances, I would expect that the collision-coalescence rate would be very small. However, the authors suggest that the aerosol size distribution is largely changed due to collection processes in Figure 5. I do not see a large change in the size distribution according to this figure.*

Reply: Thanks for your comments. We agree with your comments. We used a misleading phrase "large change" just in order to compare aerosol mass spectra in different time periods. In the new manuscript, we modified those words.

Furthermore, the series of "E" cases represent distributions with rather long tails according to Figure 1. Are the results dependent upon the tail? How important are these GCCN to the results presented in the text?

Reply: Thanks for your comments. The aerosol distribution tail of the series of "E" may impact the parameterizations just as what we mentioned in our paper that both the number concentration of aerosols and the aerosol distribution shape influence the parameterizations. However, the results of the old manuscript should not be dependent upon the tail. Actually, in order to simulate the warm rain formation, the aerosols with the diameters greater than 10um, such as these GCCN, were excluded from aerosols of such a distribution (see Figure 6b,d) (Sun et al., 2012) . In our new parameterizations, such GCCN have been included.

5. *A large portion of the analysis surrounds maximum rates. In almost any modeling framework, the maximum values of any field variable or process rate are inherently dependent upon the chosen modeling framework (i.e., dynamical core, grid resolution, time step, etc.). Very little can be learned from only examining maximum rates. I highly recommend that the authors present and analyze other statistical information regarding the processes rates.*

Reply: We appreciate these comments. We modified our manuscript as your suggestions.

6. *In general, the text is very difficult to read. I would suggest that the authors' request support from a native English speaker before providing a revised submission.*

Reply: We appreciate these comments. The new manuscript will be polished by one native English speaker.

Anonymous Referee #1

General comments:

This manuscript presents a detailed numerical study on aerosol mass conversion within water drops using a bin aerosol-microphysics scheme coupled with a 1.5-dimensional non-hydrostatic cloud model. The aerosol mass conversion rates associated with drop autoconversion and accretion processes have been examined under different initial aerosol size distributions and different radius thresholds for raindrops. The authors found that the aerosol mass conversion rate can be generally parameterized as a power-law relation to water mass coalescence rate. The exact relation and fit uncertainty (dispersion) are determined by initial aerosol size distribution. The topic of cloud processing of aerosol is appropriate for ACP in general. The work being done in this manuscript is valuable to aerosol-cloud interaction modeling community. However, the work presented here has been more or less covered by a recent study published in JAS (Lebo and Morrison, 2013). Also, some important aspects of the study are missing, which makes me hesitate to recommend publication on ACP.

Reply: We are very thankful for your fair comments. All your suggestions have been considered in our revised manuscript. We hope that the revised manuscript can be suitable for publication.

Major comments:

1.The authors mentioned several times in the manuscript that the aerosol activation is a dominant process to determine the initial cloud droplet size distribution and initial aerosol mass distribution inside drops which then determine the aerosol mass conversion process. Indeed, Lebo and Morrison (2013) also found that once a reasonably accurate aerosol activation scheme is included in a bulk scheme, the simple scaling method (aerosol mass conversion rate scales with water mass conversion rate) can simulate the cloud processing of aerosol reasonably well compared to results simulated be detailed 2-dimensional aerosol-microphysics scheme. However, there is no discussion and description of how aerosol activation is treated in this study. Given the extremely important role the aerosol activation plays in the aerosol-cloud interaction, it is necessary for the authors to explore the relation between such process and the subsequent aerosol mass conversion process in detail.

Reply: Thanks for your comments. The answers of these comments have been given for the third comment of the referee #2.

2.The case simulated here is a convective mixed-phase cloud. Although the ice-phase microphysics was turned off in the model, the warm-phase processes at high altitude are essentially unrealistic. The exclusion of ice processes will make vapor saturation ratio higher than it should be at levels below freezing point. The condensation process of water droplet is then artificially exaggerated, which impacts coalescence process and aerosol mass conversion process subsequently. Therefore, a pure warm-phase cloud case is more appropriate for the topic investigated here.

Reply: We appreciate these comments. We modified the ambient temperature lapse rate from 6 ^{o}C/km to 5.5 ^{o}C/km so that our simulations only focus on the pure warm-phase cloud case.

3.Under many conditions, the errors of the power-law relation reach an order of magnitude (Fig. 16 (g)-(i)) which indicate the uncertainties of such relations. It might be interesting to see if power-law relations of the envelop (in these cases, the majority of the points reside on the boundaries of the scatter plot) can provide better parameterization.

Reply: Thanks for your comments. In our new manuscript, the aerosol mass autoconversion rate has been parameterized as a power-law function of the aerosol mass concentration in cloud droplets, the cloud water autoconversion rate, the cloud water mass concentration and the cloud water accretion rate. The new parameterized power-law equations are much better than previous schemes.

4. The manuscript is a bit verbose. Many discussions are redundant. Significant efforts are needed to improve the readability of the manuscript.

Reply: We are very thankful for these fair comments. The new manuscript will be modified by a native English speaker.

Anonymous Referee #3

General Comments:

This study uses a 1.5-dimensional non-hydrostatic convective cloud and aerosol interaction model with bin microphysics to examine the conversion of aerosol mass in cloud droplets to aerosol mass in raindrops. The study points out that there is not linear relationship between this aerosol conversion and precipitation production as is often assumed in the parameterization of aerosol removal in global models. The study goes on to present numerous regression equations for this conversion under different aerosol size distributions and thresholds for raindrop size. The topic is appropriate for Atmospheric Chemistry and Physics but there are certain major concerns, as outlined below and as summarized by two reviewers that would need to be carefully need to be addressed before the manuscript is suitable for publication.

Reply: We are very thankful for your fair comments. We considered all your comments in our revised manuscript. We hope that the revised manuscript can be suitable for publication.

Major points:

1) Details should be provided about the activation parameterization used. The text does not address how these results might be dependent on the representation of activation and as a result how applicable these relations are to models with other activation parameterizations.

Reply: Thanks for your comments. The answers of these comments have been partly given for the third comment of the referee #2. The treatment of hygroscopic growth of aerosols has been addressed again in our new manuscript. The masses of aerosols in haze droplets and cloud droplets have been explicitly determined so that the aerosol conversion parameterizations can be applicable to any models with other activation parameterizations.

2) A large number of regression equations are provided but the text is not clear about how these are to be applied in a global model or if the authors consider such an implementation appropriate.

Reply: Thanks for your comments. The answers of these comments can be found the first comment of the referee #2.

3) The results also depend on the autoconversion and accretion parameterizations used in this study. How does this limit how broadly generalizable are the conclusions of this study, likewise for the activation scheme used? This should be explicitly addressed.

Reply: Thanks for your comments. The answers of these comments can be partly found in the second comment of the referee #2. The definition for the autoconverion and accretion of cloud droplets in the bin microphysics of our study can approximately represent the cloud droplet conversion which comes from the stochastic collision-coalescence processes. The conclusions of our study may be broadly generalizable if the aerosol size distributions are nearly the same. The point is which kind of parameterization schemes of the precipitating production rates will be applied into the aerosol-cloud interaction models with the bulk microphysics. Therefore, further studies are needed to find the best autconversion and accretion schemes of cloud droplets.

4) How does the neglect of mixed phase and ice cloud microphysics influence the conclusions of this study?

Reply: Thanks for your comments. The neglect of mixed phase and ice cloud microphysics may impact the conclusions of our study. In our new manuscript, we simulated the case without ice phase so that we can get rid of the influence of ice phase on the conclusions of our study..

5) Do you expect the results to be different for stratiform as opposed to convective clouds?

Reply: Thanks for your comments. The results from stratiform clouds without convections may be different from those obtained from convective clouds for the same aerosol size distribution because different concentrations of aerosols will be activated due to distinct supersaturations. However, stratiform clouds embedded with convections should have little difference.

6) The discussion, particularly in the Section 10 could be more concise for easier readability.

Reply: Thanks for your comments. We modified as your suggestions.

Minor Points:

Page 25500, lines 22: Should this be a reference to Table 4, not 3 here?

There are a few spelling errors , page 25505, line 11, change 'raio' to 'ratio' and page

25510, line 21, change 'remarkble' to 'remarkable'.

Reply: Thanks for your comments. We modified above errors in our new manuscript.

References:

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