We would like to thank the Referees for their constructive criticisms on our recent submission to Geoscience Model Development Discussions. They raise a number of issues, including technical questions about the new parameterization, assumptions related to the treatment of convection, and comparisons with additional measurements. We have addressed these issues in the revised version of the manuscript.

The goal of the manuscript is to provide a technical description of the new parameterization that could be utilized for a wide range of science questions related to chemical transport and the aerosol lifecycle. To document that the new parameterization is functioning as intended, we have included comparisons with data collected during the Cumulus Humilis Aerosol Processing Study (CHAPS). While development efforts remain to add additional aerosol indirect effects to the parameterization, we believe that the work described in our manuscript is the one of the first treatment of aqueous chemistry in parameterized convective clouds within WRF-Chem. As such, this work is useful to the research community at this point in time, particularly for studies related to the lifecycle of aerosol in the atmosphere. We are planning additional work to include aerosol indirect effects that will make the parameterization germane to an even wider range of questions related to climate science. We have modified the title of the manuscript and made some minor modifications to the introductory material to make these points more clear to the reader.

Both Referees commented that they would like to see more evaluation of the new parameterization with field data. We have addressed this issue by including additional *in situ* measurements from the G-1 and remote sensing measurements from the airborne NASA High Spectral Resolution Lidar (HSRL) collected during CHAPS. There are relatively few field studies that include the data needed for evaluating the impact of clouds on the aerosol population. To our knowledge, CHAPS is one of a small number of studies that included the deployment of an airborne Aerosol Mass Spectrometer (AMS) in tandem with a counter-flow virtual impactor for sampling the chemical composition of the aerosol that served as CCN. This data is necessary for some of the evaluations shown in the manuscript. We have added comparisons with the AMS data for additional days (Figure 12), as well as aerosol backscatter and extinction derived from the HSRL (Figures 7 and 8) so that we can examine changes in the vertical distribution of aerosol within the atmospheric column. It is difficult to compare the results from our simulations with other versions of WRF-Chem because most other parameterizations do not account for the impact of aqueous chemistry on aerosol, or use different chemistry packages so that a true apples-to-apples comparison is not possible. This was our original motivation for including comparisons with the high-resolutions simulations presented by Shrivastava et al. (2013), which have been removed from Section 5.2 of the revised manuscript based on the recommendations of both of the Referees.

While we acknowledge the importance of the careful evaluation of new parameterizations against field observations we would also like to point out that the aims and scope of Geoscientific Model Development, as defined on the GMD website, are:

- Geoscientific model descriptions, from box models to GCMs;

- Development and Technical papers, describing development such as new parameterizations or technical aspects of running models such as the reproducibility of results;
- Papers describing new standard experiments for assessing model performance, or novel ways of comparing model results with observational data;
- Model intercomparison descriptions, including experimental details and project protocols.

We selected GMD so that we could provide a careful description of the details of the parameterization that would be useful to the WRF-Chem user community at a level of detail that would not be possible in other peer-reviewed journals. We believe that the comparisons with data that are presented in the revised manuscript are consistent with the goals of GMD, and the work fits particularly well with the second bullet point on the list.

Referee #1 questioned our use of the Kain-Fritsch (KF) convective parameterization at the horizontal grid spacing that we used in our study. We agree with the Referee that issues related to the relevant spatial scales, parameterizations, and the model grid are often glossed over in regional scale simulations. To address this concern we have added additional text to Section 3 of the manuscript (along with an additional panel that was added to Figure 2 showing the fraction of the grid box covered by convective updrafts):

> "Care must be taken when applying cumulus parameterizations in simulations that use an intermediate grid spacing where the sub-grid scale motions can be nearly the same size as the model grid size (Wyngaard, 2004) and for cases in which the assumption that the updraft area in the model grid box is small (Arakawa et al., 2011). Alternative approaches are being developed that include new scale aware parameterizations (e.g. Gustafson et al., 2013; Grell and Freitas, 2014). In this study, the fraction of the model grid box occupied by cumulus convective updrafts was analyzed and was found to generally be less than 10% (Figure 2). The application of the cumulus parameterization at 10 km horizontal grid spacing used in this study is consistent with other work that has appeared in the literature (e.g. Larson et al., 2012; Berg et al., 2013), including Gerard et al. (2009) who identified horizontal grid spacing ranging from 2 to 7 km as problematic, and with recommendations made in the WRF Users Guide (Skamarock et al., 2008)."

Our responses to specific Referee comments are included below, highlighted in blue:

Responses to Referee 2

2652, line 12: "Preliminary testing of the modified WRF-Chem has been completed using observations from the Cumulus Humilis Aerosol Processing Study (CHAPS) as well as a high-resolution simulation that does not include parameterized convection." I don't think that "preliminary testing" is enough for a publication regarding a new cloud-aerosol treatment.

Preliminary testing was a poor word choice on our part, and we meant to say the first description and demonstration of the new parameterization. We have rephrased the sentence to more accurately reflect the status of our work.

P2654, line 24: It should be noted, however, that the modifications do not include feedbacks of aerosol on the amount of precipitation, impacts of the aerosol on the

cumulus microphysics, or feedbacks between the cumulus microphysics and the radiation. These additions are topics for subsequent research." For a new treatment of cloud-aerosol interactions, I find it crucial to have at least the feedback of aerosol on microphysics and precipitation included. Otherwise, it is hard for me to speak of cloud-aerosol "interactions". In my opinion, the authors should first implement the complete interactions, which are planned for this parameterization, before publishing.
Our work represents one of the first treatments (and the first with the Kain-Fritsch cumulus parameterization and MOSAIC aerosol chemistry) of the processing of aerosol by parameterized convective clouds in WRF-Chem and represents an important step forward even without all of the feedbacks on microphysics and precipitation. That said, we do include elements of the first aerosol indirect effect in this work (as discussed in section 5.3). The advantage of not including feedback effects is that it enables us to demonstrate the expected effects of vertical mixing and aqueous chemistry in the clouds without the complications of feedbacks.

2660, line 5: "The activation is largely a function of the cloud updraft speed." But it is also a non-negligible function of the aerosol concentration. Therefore, I am surprised that the concentration and chemical composition of aerosol particles is not included for the activation of cloud droplets.
This part of the text was misstated in the original manuscript. The idea we were trying to convey is that the updraft speed determines the activation, given a constant aerosol loading. We have modified the manuscript to make this clearer to the reader: "The activation is a function of the cloud updraft speed and the number, size, and composition of particles".

Line 12: "Once the droplet number concentrations are computed for each perturbation value of temperature and humidity in the PDF, they are averaged together to provide a single value of cloud droplet number concentration for each grid cell." Different cloud droplet number concentrations can have a significant influence on the subsequent development of a cloud. Therefore, I would imagine, that averaging the cloud droplet concentrations over the different profiles, would end up in loosing helpful information. Additionally, why are the perturbations not averaged? After that only one cloud droplet concentration has to be calculated, which would be faster concerning computational time?
The primary reason for averaging the droplet number concentrations was, as the Referee suggests, to save computational resources. The cloud droplet number concentrations that are calculated in the cumulus physics routine will eventually be used in calculations of autoconversion and cloud droplet effective radius. The shallow cumuli are non- or weakly precipitating, so the impact on autoconversion should not be very important in those clouds. The cloud drop effective radius is used by the radiation routines that (in WRF-Chem) do not treat a spectrum of clouds within a grid box. We could have averaged the perturbations of temperature and humidity together as well. That approach, however, would have some drawbacks because of non-linearities between the conditions at the cloud base and the cloud top height (due to different values of CAPE and CIN). This treatment was also selected to be consistent with the standard KF-CuP parameterization.

Line 18: “At present, secondary activation is not considered for the sub-grid convective clouds, nor does the activation feedback on the cumulus clouds via changes in the conversion of cloud water to rain” This is also an important process, when studying the interactions of aerosols and clouds. Therefore, I recommend, including this process as well in a final version of this parameterization.
We agree that secondary activation is an important process that should be included in the final version of the parameterization, and it is our intention to include it in future releases. It should be noted, however, that secondary activation is not treated for grid-resolved clouds within WRF-Chem at this point in time, and thus is not treated in high resolution (cloud-resolving) simulations of convective clouds. We have modified the text as follows: “At present, secondary activation is not considered for either sub-grid convective clouds or for high-resolution (cloud-resolving) simulations of cumulus convection…”

P2661, line 3: “passive clouds (for which the only processes are activation/resuspension and aqueous chemistry).” Earlier, it is stated that for passive clouds the vertical velocity is set to zero. Is it not inconsistent to have activation, when no updraft is present?
The Referee is correct, the word “activation” should not be in this sentence and it has been removed.

P2662, line 8: “In the cumulus-effects-on-aerosols routine, calculations are made using the properties of an average (over the population) shallow cloud, rather than doing calculations for each shallow cloud in the population.” Can the authors explain, why this is the case? What is the advantage of using an average cloud rather than a population of clouds?
This choice was made to limit the information passed between the various WRF-Chem modules and reduce the computational burden. We only average over the population for the case of shallow cumuli, and the changes in the aerosol properties associated with the cloud processes in the shallow cumuli are less sensitive to the details of the cumulus clouds. We use the full population of clouds when determining the impact of the aerosol on the cloud (where different updraft speeds can have an important impact). We have modified the text to make this more clear to the reader: “This methodology is applied to limit the information that is passed between the various WRF-Chem modules, to reduce computational burden, and to allow the same treatment for shallow and deep cumuli. The changes in aerosol properties associated with aqueous chemistry and transport in the shallow clouds are less sensitive to the details of the cumulus updrafts than is the cloud droplet number concentration.”

P2663, line 13: “Aerosol activation is calculated as described in Sect. 2.2.1, but for shallow convective clouds, the average (over different clouds) vertical velocity is used.” Since the activation is strongly dependent on the vertical velocity, is it justified to use an average vertical velocity? Would the results differ, if the an average cloud droplet concentration is calculated based on the different vertical velocities?
Activation calculations are done in both the cumulus physics routine where the focus is on the aerosol impact on the clouds, and in the cumulus chemistry routine that is used to determine the impact of the cloud on the aerosol. This duplication is due to cloud physics (including cumulus) and chemistry calculations being done in different sections of the WRF-Chem model, In the cumulus physics routine, a range of vertical velocities are

applied for shallow cumulus, based on the range of cloud properties derived from the KF-CuP treatment. In the cumulus chemistry routine, only a single representative value is used. This was done both to limit the amount of information related to the updraft parameters being passed from the physics to the chemistry routines and to reduce computational burden (e.g., avoid doing aqueous chemistry for multiple cloud profiles). The impact on simulated aerosol mass should not be large, as the cumulus updrafts are typically 1 m/s or greater, which will activate most particles of 100 nm diameter or larger, but it has some impact on aerosol number. We have made the following changes to section 2.2.1 and 2.2.2 to the manuscript, respectively:

"The activation is a function of the cloud updraft speed and the number, size, and composition of particles. In the modified version of the KF parameterization in WRF-Chem that accounts for the cloud droplet number…"

"In the cumulus-effects-on-aerosols routine, calculations are made using the properties of an average (over the population) shallow cloud, rather than doing calculations for each shallow cloud in the population. This methodology is applied to limit the information that is passed between the various WRF-Chem modules, to reduce computational burden, and to allow the same treatment for shallow and deep cumuli. The changes in aerosol properties associated with aqueous chemistry and transport in the shallow clouds are less sensitive to the details of the cumulus updrafts than is the cloud droplet number concentration."

P2663 line 26: "Cloud water can also be converted to cloud ice, but currently this is not included as part of the aerosol wet removal, as the fate of cloud ice (conversion to precipitation or detrainment near cloud top) can vary. In the future, ice processes could be incorporated in the cumulus effects routine by treating cloud-ice-borne aerosol in addition to cloud-droplet-borne aerosol." This is also a strong simplification. One could assume that for high cloud droplet concentration the formation of precipitation is delayed and hence cloud water is transformed into cloud ice. Is it justified, to neglect this effect?
This question is very similar to one raised by Referee 1, and a more detailed response is given there (under the comment about p. 2663, line 26). Note also that the conversion rate of cloud water to precipitation which is currently in the parameterization does not depend on cloud droplet number and is quite rapid. Thus the simplification currently has little impact. When extensions are made to the cloud microphysics, treating cloud-ice-borne aerosol will be more important.

Page 2665, line 24: "Because vertical velocity is assumed zero in the passive clouds, the Abdul-Razzak and Ghan (2002) parameterization cannot be used. Instead, we assume that the activated fraction for each aerosol chemical component (and size bin) is the same as the activated fraction in the steady-state updraft of the active cumulus."Why do you need activation, when the updraft is set to zero?
The Referee is correct that we would not expect new activation of particles in the passive clouds. We do, however, need to have an estimate of the number of cloud-borne particles within the passive cloud for aqueous chemistry calculations. The simplest approach is to assume that the cloud-borne fraction is the same as in the active clouds. This is consistent

with the view that the passive clouds are those that are further along in the lifecycle and were once active clouds themselves. The following change was made to the manuscript to make this more clear: "Some of the interstitial aerosol is then transferred to the convective-cloud-borne state, in order to provide an initial chemical composition of the cloud water. For this, we assume that the cloud-borne fraction for each aerosol chemical component (and size bin) is the same as the cloud-borne fraction in the steady-state updraft of the active cumulus. This is conceptually consistent with the passive clouds being decaying remnants of active clouds."

P 2676, line 5: "Some differences between the low resolution and high-resolution simulations are likely due to the averaging of the emissions over larger grid cells that produce smaller horizontal gradients in emissions that could lead to systematic differences in the aerosol loading. There are also differences in the simulated cloud field. For example, the grid-resolved simulations were free of deep convection (i.e., grid resolved clouds that one would interpret as deep convection) within the OKC analysis box while the low-resolution simulations presented here predicted a large amount of deep convection in the same box (not shown)/ These are quite significant differences. Therefore, I would rather not compare these results to Shrivastava et al. (2013) for a manuscript, which deals with the evaluation of a new model modification
Based on comments from both Referees we have removed the sections of the text that described the comparison with Shrivastava et al. (2013). Analysis of additional in situ data collected during other flight days has been added, as well as comparisons using data collected by the NASA HSRL.

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