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 1

p. 2661, line 20: "For all attachment states, the aerosol species associated with cloud droplets in the sub-grid convective clouds are treated explicitly, but only within the convective cloud routines" - A considerable fraction of precipitation in deep convection originates from the stratiform anvil region. Does the above sentence mean that once the trace gases and aerosols are detrained from the updraft, they automatically become "unattached"? Also, is uptake by cloud droplets treated in the resolved clouds? (I can not find this information in Chapman et al.) or does the detrained trace gas have to be taken up by falling hydrometeors? If yes, is this uptake kinetically limited?

The stratiform anvil is not explicitly treated in the cumulus parameterization and cloud-borne aerosol are added to the grid resolved aerosol at the altitude where detrainment from the cumulus takes place. At this point aerosol and trace gases could interact with grid-resolved anvil clouds. The manuscript has been modified to include "When air is detrained from sub-grid convective clouds, any detrained cloud-borne aerosol is added to the grid resolved interstitial aerosol in that model grid box where the aerosol can potentially interact with resolved clouds."

p. 2663, Sect. 2.2.1: "but for shallow convective clouds, the average (over different clouds) vertical velocity is used": Activation depends on the local vertical velocity, which is not the same as the multi-cloud average. The authors should at least discuss the potential impact of their assumption. They should mention whether their cumulus parameterization provides either an ensemble of updraft velocities or else assumes a PDF. In case it does neither, I would suggest to perform a sensitivity study with an assumed PDF of in-cloud updraft velocities that is consistent with the in-cloud average updraft velocity. One can expect that using the average leads to an underestimate of cloud droplet concentration, so it would be good if the authors could perform a sensitivity study on this issue.

Activation calculations are done in both the cumulus physics routine where the focus is on 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 the 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 in 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 Kain-Fritsch parameterization in WRF-Chem that accounts for the cloud droplet number, the updraft velocities associated with the buoyancy excess …"

"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."

2663, line 26: "Cloud water can also be converted to cloud ice, but currently this is not included as part of the aerosol wet removal" - Does this mean that all dissolved aerosol and trace gas is released from the hydrometeors upon freezing? If yes, this would be inappropriate for most species. In particular, several model studies have shown that releasing trace gases from hydrometeors upon freezing in deep convection strongly enhances upper tropospheric mixing ratios and reduces wet deposition.

The cloud effects on aerosols routine does not explicitly calculate or vertically transport aerosol particles attached to either cloud-ice particles or to precipitation particles (rain, snow, graupel), which is consistent with the existing grid-resolved cloud treatment described in Chapman et al. (2009). When cloud droplets are converted to or collected by precipitation particles, any aerosol material in the droplets is assumed to be immediately wet removed. When cloud droplets are converted to or collected by cloud-ice particles, the fate of the cloud-droplet-borne aerosol is left unchanged in the cumulus effects routine. So instead of becoming ice-borne, the aerosol material remains cloud-borne and moves up to the next vertical level, where it may experience more wet removal through loss to precipitation. Conceptually, this treatment is not ideal. However, the conversion of cloud water to precipitation is quite rapid in the Kain-Fritsch parameterization, so in deep clouds, most cloud-borne aerosol is wet removed before it reaches the detrainment level. If cloud-borne aerosol were to be converted to ice-borne aerosol, it would still be wet removed. We have modified the text as follows: "Cloud water could also be converted to cloud ice in the cumulus physics routine, but currently this process is not included in the aerosol wet removal calculations. The conversion rate of cloud water to precipitation that is currently used in the cumulus physics routine is quite rapid, so in deep clouds, most cloud-borne aerosol is wet removed before reaching the detrainment level, and this simplification has little impact. However, this treatment is not ideal, and 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."

p. 2663, line 6: "The environment mixing ratios for interstitial aerosol are assumed equal to the grid-cell mean values, and are zero for convective-cloud-borne aerosol." and p. 2664, line 12: "The environment gas mixing ratios are assumed equal to the grid-cell mean values" - How can this be justified at a 10 10 km2 horizontal resolution? Is the maximum updraft area fraction really small compared to the grid box size? As far as I can see, this assumption must lead to an overestimate of the uptake of trace gases. This overestimate is more severe for smaller time steps.
As shown in our general response to the Referee's comments, the convective updraft takes up a relatively small fraction of the model grid box so our assumption is defensible and use of the grid-box mean is reasonable. In addition, these environmental values are only used in determining the impact of entrainment on the cloud, and so errors associated with this assumption are likely small. We have added the following text to the manuscript near equation (6) "which is justified given the small fractional area of the grid box covered with convective updrafts."

2664, line 17: "The wet removal rate for gases only considers the removal of gases dissolved in cloud droplets; uptake of gases by rain is currently neglected." - Why?
We agree that the uptake of gases by rain should ultimately be included in treatment of cloud effects on aerosol and gases. In this work, however, we have neglected this process because of its relative importance compared to uptake in cloud. Within clouds, the wet removal associated with cloud drops is larger than raindrops due to the much larger surface area of cloud drops compared to the larger, but less plentiful, raindrops. Also, the volume of air that moves through the updraft (and experiences in-cloud wet removal) is larger than the volume that resides below cloud base but does not enter the updraft (and

experiences only below-cloud wet removal). However, future versions of the parameterizations will attempt to account for this. Our parameterization was designed with climate-relevant cloud-aerosol interactions in mind, and less attention on the wet removal of gases. The text has been modified to highlight this for the reader: "This treatment is justified within clouds because of the relatively small role of direct uptake by raindrops compared to uptake by cloud droplets followed by droplet collection by rain (due to the small surface area of raindrops compared to cloud drops). Also, the volume of air that moves through the updraft (and experiences in-cloud wet removal) is larger than the volume that resides below cloud base but does not enter the updraft (and experiences only below-cloud wet removal). Future version of the parameterization will include below-cloud wet removal."

At 10 km resolution a part of the deep convection will be resolved. Yet, the authors use a deep convection parameterization that was tailored for a lower resolution. They need to either demonstrate that this works or else find literature that demonstrates that this parameterization is appropriate for this resolution. Also: what is the maximum updraft area fraction in the 10 10 km grid boxes? As the size of the updrafts approaches the grid resolution, the assumption in the cumulus parameterization that the updraft mass flux is balanced by compensating subsidence within the same grid column breaks down. Given the potential problems with a cumulus parameterization at this resolution, one must be demonstrate that the benefits outweigh the potential problems

This question was addressed in our general responses to the Referee comments. We agree that cumulus parameterizations should be applied carefully when relatively fine model grid spacing is used. We have added additional text to the document in Section 3.1, and we have made modifications to Figure 2 showing the fraction of the grid box covered by cumulus updrafts.

Sects. 5, 5.1, and 5.2: I am not sure what can be learned from this analysis. In my opinion, it neither serves the model evaluation, nor does it contribute to an improved understanding or a better quantification of the effects of clouds on aerosols. This section should be shortened drastically. In my opinion, it does not add significantly to the existing published literature. The comparison with the high resolution simulations seems somewhat superficial, and I do not quite see what has been actually learned from it. Maybe one could try to isolate some systematic differences which could then be attributed to a cause.

Based on suggestions from Referee 1 and 2 we have made a number of modifications to section 5 of the manuscript, including removing the text dealing with the comparison of the high resolution and low-resolution simulations that had been included in Section 5.1. We have elected to keep the discussion related to the vertical distribution of aerosol on 25 June in Section 5.1.1. The section of the text devoted to the 25 June case is relatively short, and is critical to demonstrate that the new parameterization is working as expected and giving reasonable results that are consistent with our understanding of how aerosol-cloud systems work for a range of different cloud and aerosol types. To further demonstrate the parameterization is working as expected, we have also added additional analysis of the CHAPS data collected on 19 and 21 June including comparisons with the NASA HSRL to Section 5.1.1 to augment what is presented from MSN and AUS analysis boxes. We elected to keep Section 5.2 related to the regional scale impacts intact because it highlights an important use of the parameterization for regional-scale simulations. This section also highlights the relative role of aqueous chemistry in altering the aerosol loading, which is often neglected by models that do not treat aerosols within unresolved clouds.

p. 2683: “It should be noted that the current modifications do not include the treatment of other indirect effects, which will be included at a later date.” - How? Since the appropriate parameterizations do not exist, the authors should be more specific on what their plans are
The main goal of this statement was to make it clear to the reader that the treatment of many indirect effects is not included in the current version of the parameterization. We have rephrased the sentence to focus on what is included rather than what is not.

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