

## **GMD-2014-32: Response to Reviewer Comments**

We would like to thank the Reviewers and Editor for their helpful and constructive comments on the manuscript. We recognize that there are a number of physical processes missing from our parameterization (noting that many of these same processes are not included in other parameterizations), but the work we have presented is an important step in improving the treatment of cloud aerosol interactions in sub-grid convective clouds. We believe that it represents a valuable contribution to the community. From the start of the review process, we have been forthright about the processes that have been included, and those that have not. Over the course of the work, we focused on those processes that have the largest impact on the aerosol life cycle and the climate system.

At the request of the Editor, we have added additional text to the manuscript before section 2.1 (lines 13-21 on page 5) to make it absolutely clear to the reader which processes have or have not been included. In response to the comments from the second and third Reviewers, we have redrawn a number of the figures (Figures 1-7, 9, and 11), including larger labels and better shading. We have also completed a more careful investigation in regards to our use of the average vertical velocity for aqueous chemistry and transport. The second Reviewer encouraged us to provide a more quantitative estimate, but that is difficult based on the simulations that we have completed. We added additional text in Section 2.2.2 (page 11, lines 9-12 and 17-19) documenting that the median coefficient of variation was 0.11 and was less than 0.25 in 78% of the cases. Thus, the sub-grid variability in the vertical velocity is small compared to the mean value and would have a secondary impact. We have also noted that this treatment would only lead to changes in the apportionment of mass across the aerosol size bins, rather than differences in the total aerosol mass that are related to the total water and cloud fractions and the local trace gas (SO<sub>2</sub>, HNO<sub>3</sub>, etc.) mixing ratios.

## Response to Reviewer #2:

Responses to specific comments:

1. *Page 6, lines 26-28. I don't understand how the sub-grid convection can be the same proportion of the grid cell for simulations run at 100 km, 36 km, and 12 km, all of which require a cumulus parameterization. I would appreciate it if the authors could address these intermediate scales that still require a cumulus parameterization and not just jump to the < 5 km grid spacing.*

The reviewer asks how the sub-grid convection can be the same proportion of the grid box for a number of different resolutions. The solution of (1) defines the cloud fraction to be a function of two constants and the updraft mass flux. As the model grid resolution changes, the value of updraft mass flux could also change leading to variability in the cloud fraction. We have added a sentence on page 7, lines 9-11, to the text describing this performance. Additional discussion related to model resolution and convective parameterizations can be found in Section 3.1 (lines 18-29 on page 16).

2. *Page 11, lines 17-25. Is there a reason that impaction scavenging, which can occur inside the cloud and below the cloud, is not included? There are many schemes available, even with large simplification (Dana and Hales, 1976).*

We have neglected a number of processes that we deemed to be of secondary importance, and one of those is impaction scavenging of particles. We have modified the manuscript to include a list of physical processes that are not currently treated. In response to this comment, and the suggestion of the Editor, we have added text near section 2.1 on page 5 that lists processes that have not been treated in the current version of the parameterization.

3. *Page 12, line 16. Please quantify "this simplification has little impact." Is it < 10% impact when tested at  $dx = 10$  km?*

We have not quantified the impact of the assumption related to the conversion of cloud water to cloud ice. We have argued that given the simplicity of the cumulus microphysics (which essentially assumes

instantaneous removal of cloud water) that most of the activated particles are removed before the detrainment level (which we have assumed is the source of the anvil cirrus). No changes were made in the manuscript in response to this comment.

4. *Page 13, lines 8-9. On the direct uptake of gases by rain. While I agree it is a small effect, the Seinfeld and Pandis textbook address this process. It is not just a mass diffusion process to consider, but also an impaction of gases by rain.*

We agree that the uptake of gases by rain is not simply a mass diffusion process, and it is necessary to account for the fall velocity of the drop through a ventilation coefficient. In the context of this study, and as suggested by the reviewer, gas scavenging by rain is a secondary effect in these clouds (compared to the included in-cloud scavenging) and it has been neglected. In response to this comment, and the suggestion of the Editor, we have added text near section 2.1 on page 5 that lists processes that have not been treated in the current version of the parameterization.

5. *Page 15, line 9. "The net changes to the aerosol would be the same in either case". How do the authors know this? Could the differences be quantified?*

We have not attempted to quantify the differences. We have argued that the net changes would be the same because of the steady-state assumption for cloud properties over the cumulus lifetime mean that quantities such cloud water, mass flux, and cloud fractional area are constant over that time interval that the cumulus parameterization is applied. We have added an additional phrase on page 15 (lines 30-31) to make this clearer.

6. *Page 20, lines 18-20. We have known this information on aqueous-phase sulfate production since the acid rain studies of the 1980s. I suggest citing these earlier studies.*

We agree with the reviewer that the findings related to the aqueous-phase production of sulfate have been known for some time. Our intention was to highlight that the parameterization is behaving as expected. We have added the references on page 21 as suggested by the reviewer.

7. *Page 22, lines 1-2. There have been LES studies looking at tracers, simple chemistry and shallow cumulus, and gas chemistry and shallow cumulus that discuss the transport of boundary layer species to the cloud layer. It may be good to include their work in support of your findings.*

We thank the reviewer for pointing out the LES studies that are relevant to this work, and we have added the suggested references.

8. *Page 23-34. Comparisons with B200 measurements. Do you get the same results fro ShallowOnly minus Control? I find there is a lot of comparison of the DeepShllow simulation, but it is not obvious how much is from a good simulation of the shallow cumulus or from the deep convection or both. My impression is that the shallow cumulus parameterization does a good job. I don't know if the deep convection parameterization is good. Figure 8 shows that deep convection was not well simulated.*

We have completed some comparisons between the ShallowOnly and the control simulations and in response to the Reviewer's question we have added text on pages 24 and 25. The differences are subtle on 19 June, but including the deep clouds leads to a depletion of the aerosol in the boundary layer. There are differences between the results of the DeepShallow simulation for 21 June shown in Figure 8 and the ShallowOnly simulation (not shown) that demonstrates the relative importance of the deep clouds. The ShallowOnly simulation has more extinction aloft, which is associated with less wet removal and less vertical transport. This is particularly the case around 20:30 UTC, where the DeepShallow simulation has a large decrease in extinction associated with the deep clouds (**Error! Reference source not found.**). While there are no deep convective clouds shown in the lidar cross section (there were some cloud tops as high as 5 km), it is difficult to evaluate the relative skill of the parameterization to predict the occurrence of deep convective clouds from a single cross section. It should be noted that on this particular day there were number of deep clouds observed and simulated near the sampling domain.



9. *Page 26, lines 25-29. How does the ShallowOnly simulation compared (similar or different for figure 10? Perhaps a second line could be added to Figure 10b.*

We originally generated the figure using shallow clouds to be consistent with Berg et al. (2011) and Shrivastava et al. (2013) which only sampled shallow clouds in their measurements and simulations. Given that the analysis was presented for shallow clouds we do not anticipate large differences if we had analyzed the ShallowOnly simulation.

10. *Page 27, lines 17-19. We have known about the importance of nitric acid uptake and dissociation since the acid rain studies. The authors should cite these references.*

We agree with the reviewer, that the uptake of nitric acid has been known for some time. The goal of the text in the manuscript was simply to show that the parameterization is working as we had intended and that we are getting results consistent with observations.

11. *Page 27, lines 20-30. Say which simulation is discussed and why you chose to discuss those results.*

The simulations discussed in this paragraph were those of Shrivastava et al. (2013), which consisted of high-resolution simulations of shallow clouds near Oklahoma City. The intent of these sentences was simply to summarize what was presented in that paper. We have modified the text to make this clearer.

12. *Page 28, lines 30-31. Again, it is know that nitric acid is dissolved into drops and becomes part of the aerosol.*

We agree with the reviewer. The goal of the text at this stage was simply to show that the parameterization is working as we had intended and that we are getting results consistent with observations.

#### Response to clarifications

1. *Abstract. Line 13. WRF-Chem should be speed out and V3.2.1 should be added (and V3.2.1 should be removed from line 2 of page 2).*

The abstract has been modified.

2. *Page 1, line 23 does not need “as well as high-resolution simulation that does not include parameterized convection”*

The suggested change has been made.

3. *Page 5, line 14. It is awkward to say “less than 2 to 4”. Why not just say 2 to 4 km (or 1 to 4).*

While the phrasing is a bit awkward, it is not correct to say that it ranges between 2 and 4. The maximum cloud-top height for shallow convection is a function of the thermodynamics.

4. *Page 11. Line 9. What is the value of the time sub-step? Please include.*

The time sub-step is determined automatically as part of the KF-CuP parameterization and depends on the mass fluxes in a particular cloud; it is not a user specified parameter. We have modified the text to make this clearer

5. *Page 16. Line 23. Please add “subgrid” to say “impact of subgrid shallow clouds”.*

The change has been made.

6. *Page 16, last paragraph. Please clarify that grid-scale aerosol-cloud activation, cloud chemistry, and effect on autoconversion is employed in these simulations. Is it possible to do these simulations with grid-scale aerosol-cloud interactions turned off but subgrid-scale interactions turned on?*

We have modified the text to make it clear that all grid-resolved processes are turned on. It should be possible to run the simulation with the grid-resolved processes turned off, but we didn't do that in this study.

7. *Page 18, line 21. What is the “fine spatial resolution “ (dx is what value)*

The horizontal resolution used in the study of Shrivastava et al. (2013) was 2 km. We have added that to the text.

8. *Page 19, lines 9-10. Please include something like , “and the KF-CuP simulation compared better with observations than the KF only simulation.*

We have added the requested text on page 19.

9. *Page 21, line 7. Suggest writing as, “fire located in central New Mexico”*

The suggested change has been made.

10. Page 23, lines 12-13. *“due to issues with the simulated mass loading” is rather vague. I encourage the authors to be a little more specific.*

We have added some additional description on page 24 (lines 2-3).

11. Page 26, line 4. *I think it should be “cloud updraft velocity”. And CO should be defined.*

The study of Berg et al. (2011) used both updraft and downdrafts. We have modified the text to make this clearer and we have defined CO.

12. Page 28, line 10. *May be better to say “...less than was observed”*

The suggested change has been made.

13. Page 29, line 14. *I suggest not using version as it can be confused with official version of WRF-Chem.*

We have removed the word “version”.

14. Page 29, line 22. *“latter” should be “last”*

The suggested change has been made.

15. Table 1. *Could the aqueous chemistry be included with a reference?*

The table has been modified to include aqueous chemistry and associated references.

16. Table 2. *Consider having a 3 column table making it more of a matrix.*

The table has been adjusted to have three columns.

### **Response to Reviewer #3**

We have made all of the modifications to that text that were suggested by Reviewer #3.

# A New WRF-Chem Treatment for Studying Regional Scale Impacts of Cloud Processes on Aerosol and Trace Gases in Parameterized Cumuli.

L. K. Berg<sup>1</sup>, M. Shrivastava<sup>1</sup>, R. C. Easter<sup>1</sup>, J. D. Fast<sup>1</sup>, E. G. Chapman<sup>1</sup>, Y. Liu<sup>1</sup>, and R. A. Ferrare<sup>2</sup>

[1] Pacific Northwest National Laboratory, Richland, WA, USA

[2] NASA Langley Research Center, Hampton, VA, USA

Correspondence to: L. K. Berg (larry.berg@pnnl.gov)

## Abstract

A new treatment of cloud effects on aerosol and trace gases within parameterized shallow and deep convection, and aerosol effects on cloud droplet number, has been implemented in the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) version 3.2.1 that can be used to better understand the aerosol lifecycle over regional to synoptic scales. The modifications to the model include treatment of the cloud droplet number mixing ratio; key cloud microphysical and macrophysical parameters (including the updraft fractional area, updraft and downdraft mass fluxes, and entrainment) averaged over the population of shallow clouds, or a single deep convective cloud; and vertical transport, activation/resuspension, aqueous chemistry, and wet removal of aerosol and trace gases in warm clouds. These changes have been implemented in both the WRF-Chem chemistry packages as well as the Kain-Fritsch cumulus parameterization that has been modified to better represent shallow convective clouds. Testing of the modified WRF-Chem has been completed using observations from the Cumulus Humilis Aerosol Processing Study (CHAPS). The simulation results are used to investigate the impact of cloud-aerosol interactions on regional scale transport of black carbon (BC), organic aerosol (OA), and sulfate aerosol. Based on the simulations presented here, changes in the column integrated BC can be as large as -50% when cloud-aerosol interactions are considered (due largely to wet removal), or as large as +40% for sulfate in non-

precipitating conditions due to sulfate production in the parameterized clouds. The modifications to WRF-Chem are found to account for changes in the cloud drop number concentration (CDNC) and changes in the chemical composition of cloud-drop residuals in a way that is consistent with observations collected during CHAPS. Efforts are currently underway to port the changes described here to the latest version of WRF-Chem, and it is anticipated that they will be included in a future public release of WRF-Chem.

## **1 Introduction/motivation**

There remains a significant amount of uncertainty related to both the aerosol direct forcing and aerosol indirect effects (e.g. Solomon et al., 2007; Boucher et al., 2013). Numerical models of the atmosphere are one of the common tools used to investigate these effects. High-resolution simulations using horizontal grid spacing less than 10 km, which can explicitly represent convective clouds and cloud-aerosol interactions, have been widely used for short-term studies of cloud-aerosol interactions (e.g. Qian et al., 2009; Wang et al., 2011; Fan et al., 2012). They have not, however, generally been used for long-term simulations because of the associated computational expense. For long-term simulations, coarser horizontal resolution is generally required that necessitates the use of a cumulus parameterization even if the cloud-aerosol interactions associated with sub-grid scale convective clouds are poorly represented (e.g. Zhao et al., 2011). Thus, treatments of aerosols in cumulus parameterizations are needed for investigations of the impact of clouds on aerosol mixing, transformation, and removal as well as the impact of aerosol on cloud properties (Stevens and Feingold, 2009).

Shrivastava et al. (2013) compared changes in the aerosol chemical composition and cloud microphysical structure associated with cloud-aerosol interactions in fields of shallow cumuli to data collected during the Cumulus Humilis Aerosol Processing Study (CHAPS; Berg et al., 2009). The main goal of CHAPS was to find evidence of cloud-aerosol interactions in fields of shallow cumuli. The simulations presented by Shrivastava et al. (2013) were completed with sufficiently high resolution that a convective parameterization was not required allowing them to investigate cloud-aerosol interactions

1 in relatively small shallow clouds that would be sub-grid scale at coarser resolutions.  
2 Among their findings were systematic changes in the chemistry of activated particles and  
3 cloud microphysics within shallow cumuli. They found that nitric acid vapor uptake by  
4 cloud droplets led to increased nitrate content in the cloud droplet residuals. They also  
5 reported changes in cloud microphysical properties, with increases in cloud droplet  
6 number concentration and decreases in droplet effective radius with an increase in  
7 pollutant loading.

8 The Weather Research and Forecasting model coupled with Chemistry (WRF-Chem)  
9 (Grell et al., 2005; Fast et al., 2006) is frequently used to simulate conditions over a range  
10 of spatial scales and has been used to study a wide range of atmospheric phenomena  
11 associated with atmospheric chemistry and aerosols (e.g. McKeen et al., 2007; Ntelekos  
12 et al., 2009; Grell et al., 2011; Pfister et al., 2011; Ahmadov et al., 2012; Matsui et al.,  
13 2013). To date, however, the treatment of cloud-aerosol interactions has largely been  
14 limited to grid-resolved clouds, **which can be convective clouds if the model resolution is**  
15 **sufficiently fine (e.g. Chapman et al., 2009; Saide et al., 2012; Zhang et al., 2013;**  
16 **Eidhammer et al., 2014; Mashayekhi and Sloan, 2014).** This is the case for WRF coupled  
17 with the Community Atmospheric Model version 5 (CAM5; Neale et al., 2012) physics  
18 parameterizations, although cloud-aerosol interactions in convectively detrained  
19 stratiform clouds are treated (Ma et al., 2013). One exception is the recent modification  
20 of the Grell cumulus parameterization (Grell, 1993; Grell and Dévényi, 2002) to include  
21 aqueous chemistry using Community Multiscale Air Quality (CMAQ) routines and  
22 aerosol interactions in the conversion of cloud water to rainwater and the evaporation of  
23 rain (Grell and Freitas, 2014). Lim et al. (2013) added a treatment of aerosol activation to  
24 the Zhang-McFarlane parameterization (Zhang and McFarlane, 1995) while Zhao et al.  
25 (2013) modified the Kain Fritsch scheme to better account for transport and wet  
26 scavenging of dust, but each of their modifications do not include treatment of aqueous  
27 chemistry in the clouds nor have they been added to the publicly released version of  
28 WRF-Chem. To address this missing process, we have modified WRF-Chem to include  
29 treatments of a number of factors and processes important for accurately representing  
30 aerosol and trace gases within sub-grid convective clouds, including: fractional coverage  
31 of active and passive clouds, vertical transport, activation and resuspension, wet removal,

1 and aqueous chemistry for cloud-borne particles. The new parameterization uses the  
2 Model for Simulating Aerosol Interactions and Chemistry (MOSAIC; Zaveri et al., 2008)  
3 packages to represent the aerosol chemistry. This new treatment is important to include  
4 additional realism in regional scale modeling studies that require the use of cumulus  
5 parameterizations when investigating the effects of clouds on aerosol and the effects of  
6 aerosol on clouds. It should be noted, however, that the modifications do not yet include  
7 feedbacks of aerosol on the amount of precipitation, or feedbacks between the cumulus  
8 microphysics and the radiation. These additions are topics for subsequent research.

9 The work presented here describes the implementation of a treatment of activation,  
10 vertical transport, aqueous chemistry, and wet removal for sub-grid parameterized  
11 convective clouds in WRF-Chem. Section 2.0 describes changes to both the standard  
12 cumulus parameterization and the treatment of processes affecting aerosol and trace gases  
13 in the sub-grid convective clouds. These changes include improved treatment of cloud  
14 fraction as well as treatment of cloud droplet number concentration, vertical transport,  
15 activation/resuspension, aqueous phase chemistry, and wet removal. Section 3 provides a  
16 description of the WRF-Chem configuration, simulation design, and emissions data used  
17 in the study. The data used from CHAPS are presented in Section 4. Analysis of the  
18 WRF-Chem simulations are presented in Section 5. Rather than focusing on only the  
19 CHAPS study area, results are also presented from three different locations that were  
20 selected to highlight the performance of the model in situations with shallow and deep  
21 sub-grid convective clouds; and to document the impact on the regional scale transport,  
22 cloud microphysics, and the chemical composition of cloud droplets.

## 23 **2 Modifications to WRF-Chem**

24 The primary goal of this effort has been to improve the representation of vertical  
25 transport, aqueous chemistry, wet removal, and aerosol effects on cloud droplet number  
26 in parameterized sub-grid convective clouds within WRF-Chem. To address this goal, a  
27 number of modifications shown schematically in Figure 1, have been made to WRF-  
28 Chem in order to account for cloud-aerosol interactions within these clouds. These  
29 modifications include changes to the Kain-Fritsch (KF) cumulus scheme (Kain and  
30 Fritsch, 1990; Kain, 2004) as well as changes designed to account for transport,

1 transformation, and removal of aerosols and trace gases within sub-grid convective  
2 clouds.

3 The WRF-Chem model architecture separates physical processes involving sub-grid  
4 cumulus, microphysics for grid-resolved clouds, boundary-layer turbulence, and radiation  
5 from processes involving aerosol and trace gases. We have followed this separation, so  
6 that code changes involve both a cumulus physics routine that determines the presence of  
7 sub-grid convective clouds, their properties, and their impacts on heat, moisture, and  
8 momentum, and a separate cumulus aerosol and trace gas routine that treats vertical  
9 transport, activation/resuspension, aqueous chemistry, and wet removal of aerosol and  
10 trace gases. Modifications to the cumulus physics routine are described in sections 2.1.1,  
11 2.1.2, and 2.2.1. The cumulus aerosol and trace gas routine, which is new to WRF-Chem,  
12 is described in section 2.2.2.

13 The parameterization described in this work is an important first step in developing  
14 improved treatments of cloud-aerosol interactions and aqueous chemistry in sub-grid  
15 convective clouds. As such, the focus has been on capturing the first order effects  
16 necessary for representing cloud-aerosol interactions and their impact on the aerosol  
17 lifecycle. For this reason a number of secondary processes were not incorporated during  
18 this first step, including: direct scavenging of trace gases by rain, impaction scavenging  
19 of particles by rain, secondary activation, the effect of changing cloud drop number  
20 concentration on radiation, and the second indirect effect. Each of these processes,  
21 however, can be added at a later date.

## 22 **2.1 Modifications to the Kain-Fritsch cumulus parameterization**

### 23 **2.1.1 Trigger function for convection**

24 Recently, the KF scheme has been modified to improve the treatment of shallow cumuli,  
25 which are defined by the KF scheme to be less than 2 to 4 km in height, depending on the  
26 temperature at the lifting condensation level. These changes were made primarily within  
27 the standard KF (Kain and Fritsch, 1990; Kain, 2004) convective parameterization and  
28 involved replacing the default ad-hoc trigger function used in the parameterization with  
29 one explicitly linked to the boundary-layer turbulence. This was accomplished using the



Cumulus Potential (CuP) scheme (Berg and Stull, 2005) leading to the new KF-CuP parameterization (Berg et al., 2013). These changes were designed to better account for sub-grid variability by applying a range of temperature and moisture perturbations from the grid-box mean as the convective trigger, thus allowing a population of shallow clouds with different thermodynamic properties to coexist in a model grid column. In the case of deep convection, only the single most probable temperature and moisture perturbation that triggered clouds is applied to be consistent with the standard implementation of the KF scheme. The sub-grid distribution of temperature and humidity was parameterized using probability density functions (PDFs) of temperature and humidity that were based on the jump of potential temperature and moisture at the surface and at the boundary-layer top (Berg and Stull, 2004). These modifications, along with the cloud fraction changes (Section 2.1.2), were shown to significantly increase the frequency of occurrence of simulated shallow clouds over the Southern Great Plains, leading to improved forecasts of both cloud fraction and downwelling shortwave irradiance (Berg et al., 2013). It should also be noted that while the new trigger function is not scale aware, it could easily be modified to adjust the PDF based on the model grid spacing.

### 2.1.2 Cloud fractional area

In their modifications to the standard KF scheme, Berg et al. (2013) included a simple treatment of the cloud fraction associated with sub-grid scale convective clouds. Their method was based on representative time scales associated with cumulus, which the method defined to be a function of the cloud depth, turbulence intensity, and the moisture in the cloudy layer. In the work presented here, an additional treatment was added to determine the cloud fractional area for instances with deep convection. Rather than develop a new representation of the total cloud fraction for deep convection the empirical treatment used in CAM5 is applied. In this parameterization the cloud fraction associated with deep convection is a function of the convective mass flux (Neale et al., 2012), and is represented as:

$$\sigma_{dp,cu} = k_{1,dp} \ln(1 + k_2 M_{dp,cu}), \quad (1)$$

1 where  $\sigma_{dp,cu}$  is the cloud fraction associated with deep-convective clouds,  $k_{1,dp}$  is an  
 2 adjustable parameter set to 0.1,  $k_2$  is assumed to be 675, and  $M_{dp,cu}$  is the updraft mass  
 3 flux of the convective clouds (in  $\text{kg m}^{-2} \text{s}^{-1}$ ). The values of both  $k_{1,dp}$  and  $k_2$  were selected  
 4 to be the same as the values used in CAM5, and are identical to those used by Ma et al.  
 5 (2013) in their implementation of the CAM5 physics in WRF and made publically  
 6 available in version 3.5 and are similar to that proposed by Alapaty et al. (2012) and  
 7 Herwehe et al. (2014). It could be argued that a parameterization of cloud fraction  
 8 developed for a relatively coarse resolution model like CAM5 is not appropriate for a  
 9 regional scale model like WRF, which can be run at a wide range of resolutions. **The**  
 10 **cloud fraction defined by (1) can vary with model grid spacing due to changes associated**  
 11 **with  $M_{dp,cu}$ , and as such has some scale dependence.** When run at high horizontal  
 12 resolution, however, the cumulus parameterization is generally turned off so that the  
 13 parameterization of sub-grid convective cloud fraction is not utilized. Given the constants  
 14 define above, (1) predicts the maximum cloud fraction in the grid cell associated with  
 15 deep convection to be approximately 45%. Similar to the methodology applied by Berg et  
 16 al. (2013) for cases of shallow cumuli, the deep-cloud fraction computed using (1) is  
 17 applied in the radiation parameterization but has no impact on either the convective  
 18 tendencies for heat, moisture, momentum or on the cumulus transport of aerosols and  
 19 trace gases. It is, however, used in the computations related to aqueous chemistry  
 20 described in Section 2.2.

21 The cloud fraction associated with both shallow and deep sub-grid convective clouds is  
 22 broken further into two sub-types: active and passive clouds (e.g. Stull, 1985). Active  
 23 clouds are those that have vigorous updrafts and contribute to the upward cloud mass  
 24 fluxes. The fractional area of active cumulus for shallow clouds is defined as the fraction  
 25 of the PDF of temperature and humidity applied in the convective scheme that forms  
 26 clouds, while for deep clouds it is the standard KF updraft fraction area. Passive clouds  
 27 consist primarily of decaying clouds without a well-organized updraft. The fractional area  
 28 of the passive clouds is determined as the difference between the total cloud fraction  
 29 [computed following Berg et al. (2013) for shallow clouds, and Eq. 1 for deep clouds]  
 30 and the active cloud fraction that is determined within the KF-CuP scheme. Passive  
 31 clouds are treated as quiescent and are assigned zero vertical velocity, so that there is no

vertical mass flux. They are assumed to have the same total cloud water and ice content as the active clouds, but be non-precipitating, so there is no wet removal associated with passive clouds. In addition, when a convective cloud is triggered in a model grid column, the cloud population is assumed to be in steady state over the cloud lifetime defined in the cumulus parameterization (30 minutes for shallow clouds and 30 to 60 minutes for deep clouds).

## **2.2 Modifications to account for vertical transport, aqueous chemistry, wet removal, and cloud droplet number**

Chapman et al. (2009) described a treatment of cloud-aerosol interactions for grid resolved clouds within WRF-Chem. For cloudy grid cells, the standard version of WRF-Chem treats both cloud borne (activated) and interstitial (nonactivated) particles as separate transported species. A number of modifications to the standard WRF-Chem version 3.2.1 have been implemented in this study to specifically address cloud-aerosol interactions in sub-grid convective clouds. These modifications include calculations for:

- Cloud droplet number mixing ratio
- Cloud microphysical (conversion rates, and cloud water and cloud ice mixing ratios) and cloud macrophysical properties (updraft fractional area, updraft and downdraft mass fluxes, and entrainment) averaged over the population of shallow convective clouds, or for the single deep convective cloud value, and
- Vertical transport, activation/resuspension, aqueous chemistry, and wet removal of aerosols and trace gases.

WRF-Chem has several different aerosol and trace gas representations, which are referred to as chemistry packages. Our changes for sub-grid convective clouds were implemented with the MOSAIC (Zaveri et al., 2008) sectional aerosol model and the SAPRC-99 photochemical mechanism (Carter, 2010). Extension to other WRF-Chem chemistry packages would be relatively straightforward, especially those packages for which aqueous chemistry and aerosol activation modules (or interfaces) already exist.

### **2.2.1 Aerosol effects on the cloud droplet number**

Within the default KF scheme, as well as other cumulus parameterizations applied in WRF, a highly simplified treatment of cloud microphysics is used. Cloud water is produced in updrafts and converted to precipitation based on a prescribed e-folding height, and additional assumptions are made involving frozen condensate and precipitation and detrainment to downdrafts (e.g. Kain and Fritsch, 1990). While such a simplified treatment has been successful for mesoscale weather forecasting, it is not sufficient for studying cloud-aerosol interactions that are intimately linked to the cloud microphysics. Thus, the activation of cloud drops in convective drafts must be considered. 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, the updraft velocities associated with the buoyancy excess are computed using the temperature and humidity perturbations for the range of parcels identified by the KF-CuP parameterization that form clouds. Further, the droplet activation for each perturbation is computed by applying an entraining parcel conceptual model using the Abdul-Razzak and Ghan (2000) parameterization modified to account for entrainment following Barahona and Nenes (2007). 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. Above cloud base, the number of cloud droplets is further reduced by entrainment, where the entrainment rates are determined using the KF scheme (averaged over all of the parcel perturbations to yield a single entrainment rate). At present, secondary activation is not considered for either sub-grid convective clouds or for high-resolution (cloud-resolving) simulations of cumulus convection. In addition, the activation does not feedback on the cumulus clouds via changes in the conversion of cloud water to rain (as treated by Grell and Freitas, 2014).

### **2.2.2 Effects of sub-grid cumulus on aerosol and trace gases**

A new module was introduced to WRF-Chem to calculate the effects of sub-grid convective clouds on aerosol and trace gases, including vertical transport,

1 activation/resuspension, aqueous chemistry in cloud droplets, and wet removal. The new  
2 module has separate sections that treat the active clouds (as well as vertical transport in  
3 the subsiding environment surrounding the active clouds) and passive clouds (for which  
4 the only process is aqueous chemistry).

5 In models of the cloud (and precipitation) effects on aerosols and trace gases, one must  
6 consider the attachment state (Ghan and Easter, 2006) of (aerosol) particles and gases.  
7 For example, interstitial aerosol particles (i.e., particles suspended in air) may become  
8 attached to, dissolved in, or suspended in various hydrometeors (cloud and rain drops, ice  
9 crystals, snow and graupel particles). When the aerosol representation involves several  
10 size bins (8 in our study) and multiple chemical species within each bin (14 in our study),  
11 the computational expense of explicitly treating all possible attachment states is  
12 considerable, and simplifying assumptions are often used. For example, in Chapman et al.  
13 (2009) the treatment of cloud-aerosol interactions focused on grid-resolved warm clouds.  
14 Aerosol material (sulfate, nitrate, etc.) associated with cloud droplets (referred to as  
15 cloud-borne) of grid-resolved clouds was treated explicitly as transported prognostic  
16 species, while moderately soluble gases dissolved in cloud droplets were assumed to be  
17 in equilibrium with the gas-phase and were treated diagnostically. Aerosol material and  
18 gases that became associated with precipitation particles (rain, snow, graupel) and also  
19 ice crystals were assumed to be quickly removed from the atmosphere and were not  
20 treated explicitly. A similar but somewhat simpler approach is used in our treatment of  
21 sub-grid cumulus effects. For all attachment states, the aerosol species associated with  
22 cloud droplets in the sub-grid convective clouds are treated explicitly, but only within the  
23 convective cloud routines. This approximation is reasonable because of the relatively  
24 short life-time of the parameterized convective clouds (30-60 minutes) and the fact that  
25 the parameterization is intended for use with model horizontal grid spacings of  
26 approximately 10 km or more. When air is detrained from sub-grid convective clouds,  
27 any detrained cloud-borne aerosol is added to the grid resolved interstitial aerosol in that  
28 grid box where the aerosol can potentially interact with resolved clouds.

29 The cumulus physics routine determines if sub-grid convective cloud is present within a  
30 model grid column and the physical properties of the cumulus clouds (shallow or deep;  
31 life-time; updraft and downdraft mass fluxes, entrainment, and vertical velocity; mixing

ratios of cloud water, ice, and precipitation; and microphysical conversion of cloud water to cloud ice and precipitation) that are used in the cumulus effects routine. Within the KF-CuP scheme, when deep convection is diagnosed within a grid column, the deep clouds are assumed identical, and there is a single vertical profile for updraft and downdraft mass fluxes and each microphysical parameter. When shallow convection is diagnosed, there is a population of shallow clouds with different profiles, and downdrafts are not treated. 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. In this study, the median coefficient of variability of the cloud-base vertical velocity for shallow sub-grid convective clouds was 0.11, and 78% of all values were less than 0.25, which highlights that the variability in the updraft strength within a grid cell is relatively small. 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 population of cumulus updrafts than is the cloud droplet number concentration, and the largest impact is on the distribution of aerosol mass between the size bins (as determined by which bins are activated) rather than changes in the total aerosol mass.

Active cloud calculations are performed first, followed by passive cloud calculations. The treatment of active sub-grid cumulus effects on aerosols and gases is very similar to the unified treatment described in the supplementary material of Wang et al. (2013). The active-cloud updrafts and downdrafts are treated as steady-state entraining plumes. The updraft and downdraft mass fluxes obey

$$\frac{\partial M_Y}{\partial z} = \frac{(E_Y - D_Y)}{\Delta z} \quad (2)$$

where the  $Y$  subscript is either  $U$  for updraft or  $D$  for downdraft,  $M_Y$  is the mass flux ( $\text{kg m}^{-2} \text{ s}^{-1}$ ) defined at vertical layer boundaries, and  $E_Y$  and  $D_Y$  are the entrainment and detrainment in a layer, and  $\Delta z$  is the layer thickness. The compensating mass flux in the environment,  $M_E$ , is equal to  $-(M_U + M_D)$ . The active-cloud calculations involve

integrating conservation equations for grid-cell mean mixing ratios of aerosol and trace gas species over the lifetime of the cumulus cloud. The integration uses an internal time sub-step selected automatically so that the transport of air out of a layer (by  $M_E$ ,  $E_U$ , and  $E_D$ ) during the sub-step does not exceed the layer's air mass  $\rho\Delta z$ , where  $\rho$  is the air density.

For each time sub-step, steady-state vertical profiles of aerosol and trace gas species in the updraft and downdraft are first calculated. This is done by integrating steady-state continuity equations upwards (for updrafts) or downwards (for downdrafts). For aerosol species in the updraft, the continuity equation is

$$\frac{\partial(M_U q_{X,U})}{\partial z} = \frac{(E_U q_{X,E} - D_U q_{X,U})}{\Delta z} + \rho A_U \left[ (\dot{q}_{X,U})_{ACTI} + (\dot{q}_{X,U})_{WETR} + (\dot{q}_{X,U})_{AQCH} \right] \quad (3)$$

Here  $q_{X,E}$  and  $q_{X,U}$  are aerosol mixing ratios in the environment ( $E$ ) and updraft ( $U$ ), respectively, the  $X$  subscript is either  $AI$  for interstitial aerosol species or  $ACC$  for convective-cloud-borne (activated) aerosol species. The environment mixing ratios for interstitial aerosol are assumed equal to the grid-cell mean values, and are zero for convective-cloud-borne aerosol.  $A_U$  is the updraft fractional area and is equal to  $(M_U/\rho w_U)$ , where  $w_U$  is the updraft vertical velocity. The last three terms on the right hand side are the rates of change due to activation ( $ACTI$ ), in-cloud wet removal ( $WETR$ ), and aqueous-phase chemistry within cloud droplets ( $AQCH$ ). For interstitial aerosol, only the activation term is non-zero.

Aerosol activation is calculated as described in section 2.2.1, but with the simplification of using the average (over different clouds) vertical velocity for shallow cumuli rather than range of values that is used in the cumulus physics routine (the reasons for this simplification were discussed earlier in this sub-section). The Abdul-Razzak and Ghan (2000) parameterization provides activation fractions ( $f_{ACT}$ ) for aerosol number and mass species in each size bin. The activation rate in (3) is then

$$(\dot{q}_{ACC,U})_{ACTI} = -(\dot{q}_{AI,U})_{ACTI} = (f_{ACT} q_{AI,U})/\Delta t_U \quad (4)$$

where  $\Delta t_U = \Delta z/w_U$  is the time for updraft air to move across a layer.

The wet removal rate for cloud-borne aerosol in (3) is given by

$$1 \quad (\dot{q}_{ACC,U})_{WETR} = -(f_{WETR} q_{ACC,U}) / \Delta t_U \quad (5)$$

2 where  $f_{WETR}$  is the fractional removal of cloud-borne aerosols in the updraft as they move  
 3 across a layer. This fractional removal is currently equal to the fractional conversion of  
 4 cloud-water to precipitation across the layer, which is provided by the cumulus physics  
 5 routine. Cloud water could also be converted to cloud ice in the cumulus physics routine,  
 6 but currently this process is not included in the aerosol wet removal calculations. The  
 7 conversion rate of cloud water to precipitation that is currently used in the cumulus  
 8 physics routine is quite rapid, so in deep clouds, most cloud-borne aerosol **are** wet  
 9 removed before reaching the detrainment level, and this simplification has little impact.  
 10 However, this treatment is not ideal, and in the future, ice processes could be  
 11 incorporated in the cumulus effects routine by treating cloud-ice-borne aerosol in  
 12 addition to cloud-droplet-borne aerosol.

13 The aqueous-phase chemistry rate in (3) is obtained by calling the WRF-Chem cloud-  
 14 chemistry routine for grid-resolved clouds (Chapman et al., 2009). This routine calculates  
 15 mixing ratio changes from gas uptake and aqueous-phase reactions in an air parcel (or  
 16 layer) over a specified time step, and it is applied to updraft air moving across a layer in  
 17 time  $\Delta t_U$ .

18 For trace gases in the updraft, the continuity equation is

$$19 \quad \frac{\partial(M_U q_{G,U})}{\partial z} = \frac{(E_U q_{G,E} - D_U q_{G,U})}{\Delta z} + \rho A_U [(\dot{q}_{G,U})_{WETR} + (\dot{q}_{G,U})_{AQCH}] \quad (6)$$

20 where  $q_{G,E}$  and  $q_{G,U}$  are gas mixing ratios in the environment and updraft, respectively.  
 21 The environment gas mixing ratios are assumed equal to the grid-cell mean values, which  
 22 is justified given the small fractional area of the grid box covered with convective  
 23 updrafts. The  $q_{G,U}$  includes both gas-phase and dissolved in convective cloud-water  
 24 species (e.g., gaseous  $SO_2$  plus S(IV) in cloud water). The WRF-Chem cloud-chemistry  
 25 routine gives the aqueous-phase chemistry rate in (6), as well as the fraction of the gas  
 26 that is dissolved in convective cloud-water ( $f_{G,CCW}$ ). The wet removal rate for gases only  
 27 considers the removal of gases dissolved in cloud droplets; and direct uptake of gases by  
 28 rain is currently neglected. 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. The wet removal rate in (6) is then

$$(\dot{q}_{G,U})_{WETR} = -(f_{WETR} f_{G,CCW} q_{G,U}) / \Delta t_U . \quad (7)$$

Downdrafts are assumed to be sub-saturated and contain no cloud droplets or convective-cloud-borne aerosol. Thus activation, wet removal, and aqueous-phase chemistry are not treated in downdrafts. The downdraft continuity equations are then

$$\frac{\partial(M_D q_{X,D})}{\partial z} = \frac{(E_D q_{X,E} - D_D q_{X,D})}{\Delta z} \quad (8)$$

where  $X$  is either  $AI$  for interstitial aerosol species or  $G$  for gases.

Once the aerosol and gas profiles in the updraft and downdraft have been calculated, conservation equations for grid-cell mean mixing ratios of aerosol and trace gas species are integrated for the time sub-step. These conservation equations have the form

$$\rho \frac{\partial \bar{q}_X}{\partial t} = -\frac{\partial}{\partial z} [M_U q_{X,U} + M_D q_{X,D} + M_E q_{X,E}] + \rho A_U [(\dot{q}_{X,U})_{ACTI} + (\dot{q}_{X,U})_{WETR} + (\dot{q}_{X,U})_{AQCH}] \quad (9)$$

where the  $X$  subscript is either  $AI$ ,  $ACC$ , or  $G$ , and the updraft rate of change terms come from the updraft calculations described above. The integration is explicit in time and uses simple upstream finite differencing for the vertical transport terms. After the integration sub-step, the grid-cell mean mixing ratio of convective-cloud-borne aerosol ( $\bar{q}_{ACC}$ ) may be non-zero at or near levels where the updraft detrains. This convective-cloud-borne aerosol is partially transferred to grid-resolved cloud-borne aerosol (fraction transferred equal to grid-resolved cloud fraction) and partially resuspended to interstitial aerosol. At the end of all the active-cloud integration sub-steps, the new grid-cell mean aerosol and gas mixing ratios reflect the effect of the active cumulus cloud over the cloud lifetime.

1 The passive cumulus effects calculations are performed next. These calculations are  
2 relatively simple in comparison, as there is no vertical transport or wet removal of  
3 aerosol. The cumulus physics routine provides the passive cumulus cloud fraction and  
4 cloud water mixing ratio at each vertical level. Initial mixing ratios of interstitial aerosol  
5 and trace gases are set equal to the grid-cell mean mixing ratios at the end of the active  
6 cumulus effects calculation. Some of the interstitial aerosol is then transferred to the  
7 convective-cloud-borne state, in order to provide an initial chemical composition of the  
8 cloud water. For this, we assume that the cloud-borne fraction for each aerosol chemical  
9 component (and size bin) is the same as the cloud-borne fraction in the steady-state  
10 updraft of the active cumulus. This is conceptually consistent with the passive clouds  
11 being decaying remnants of active clouds. Aqueous-phase chemistry calculations are then  
12 made for this passive cloud fraction, again over the lifetime of the cumulus. Finally, the  
13 passive cumulus fraction of the grid cell is mixed with the remainder of the grid cell, and  
14 convective-cloud-borne aerosol is partially transferred to grid-resolved cloud-borne and  
15 partially resuspended to interstitial.

16 After the passive cloud calculations, the grid-cell mean mixing ratios of aerosols and  
17 trace gases reflect the effects of active and passive cumulus over the cloud lifetime.  
18 These mixing ratios are returned to the host code as the updated mixing ratios. In our  
19 simulations, a primary time step (for dynamics) of 15 seconds was used, and a chemistry  
20 time step (for most processes involving trace gases and aerosols) of 5 minutes was used.  
21 The sub-grid cumulus lifetimes, as defined within the cumulus parameterization, ranged  
22 between 30 and 60 minutes, and the cumulus effects on aerosols/gases are calculated  
23 once only when a cumulus is triggered in a grid column. On subsequent chemistry time  
24 steps, no more cumulus effect calculations are performed until a new cumulus is triggered  
25 in a column. An alternate approach would be to save the cumulus effects tendencies for  
26 aerosols and gases, then apply them gradually over the cumulus lifetime, analogous to the  
27 approach used in the cumulus physics for temperature, moisture, and momentum. We  
28 chose this one-time update approach for aerosols and gases for simplicity and to reduce  
29 memory costs associated with storing the cumulus effects tendencies for the many aerosol  
30 and gas species. The net changes to the aerosol would be the same in either case **because**  
31 **of the steady-state assumption used for the cloud properties over the cumulus lifetime,**

1 but the changes are applied somewhat sooner in the once-only approach (when a cloud  
2 triggers rather than over its lifetime), producing small differences in a simulation that  
3 could grow over time.

### 4 **3 WRF-Chem configuration**

#### 5 **3.1 Experiment setup**

6 WRF-Chem version 3.2.1 was configured in a way similar to that described by  
7 Shrivastava et al. (2013). A single domain, 2240 km on a side, over the central United  
8 States was used with 10 km horizontal grid spacing. WRF-Chem was also configured to  
9 use 64 vertical levels, with approximately 25 levels in the lowest 1 km of the atmosphere.  
10 The various parameterizations utilized in the simulations, not including the modifications  
11 described in Section 2, are listed in Table 1. Multi-day WRF-Chem simulations for the  
12 period of 1 June through 30 June 2007 were completed in individual 36-hour blocks. The  
13 first 12 hours of each block were discarded and the final 24 hours saved for analysis.  
14 Meteorological initial and boundary conditions for each block were taken from the  
15 Global Forecast System (GFS). Boundary conditions of trace gases and aerosols were  
16 derived from the MOZART global simulation (Emmons et al., 2010b). Initial conditions  
17 for trace gases and aerosol were taken from the end of the previous simulation block.

18 Care must be taken when applying cumulus parameterizations in simulations that use an  
19 intermediate grid spacing where the sub-grid scale motions can be nearly the same size as  
20 the model grid size (Wyngaard, 2004) and for cases in which the assumption that the  
21 updraft area in the model grid box is small (Arakawa et al., 2011). Alternative approaches  
22 are being developed that include new scale aware parameterizations (e.g. Gustafson et al.,  
23 2013; Grell and Freitas, 2014). In this study, the fraction of the model grid box occupied  
24 by cumulus convective updrafts was analyzed and was found to generally be less than  
25 10% (Figure 2). The application of the cumulus parameterization at 10 km horizontal grid  
26 spacing used in this study is consistent with other work that has appeared in the literature  
27 (e.g. Larson et al., 2012; Berg et al., 2013), including Gerard et al. (2009) who identified  
28 horizontal grid spacing ranging from 2 to 7 km as problematic, and with  
29 recommendations made in the WRF Users Guide (Skamarock et al., 2008).

Three sets of simulations are used to investigate the regional impacts of cloud-aerosol interactions associated with both shallow and deep convection (Table 2). In all three simulations, the shallow and deep cumulus physics are enabled as well as aerosol processes (activation/resuspension, aqueous chemistry, and removal) in grid-resolved clouds. However, the cumulus effects on aerosols and trace gases are selectively enabled in the different simulations. The first simulation includes aerosol processing associated with both shallow and deep clouds, and is referred to as DeepShallow. This simulation can be used to estimate the regional impact on aerosol properties due to cloud processing associated with all clouds in the domain (including both grid resolved and parameterized clouds). The second simulation has aerosol processing by shallow convection turned on and by deep convection turned off, and is referred to as ShallowOnly. The difference between DeepShallow and ShallowOnly is used to document the impact of aerosol processing by deep convection alone and is identified as the Deep-Effect in this work. The third simulation is conducted with all aerosol processing by sub-grid convective clouds turned off (Control) and is the default treatment in WRF-Chem. The difference between ShallowOnly and Control simulations show the impact of sub-grid shallow clouds and will be identified as the Shallow-Effect in the rest of the manuscript. An additional simulation was completed for a subset of the study period to document the impact of aqueous phase cloud chemistry on aerosol composition. This was accomplished by repeating the DeepShallow simulation for 25 June 2007 with the convective cloud aqueous chemistry turned off. This run was initialized using the aerosol from the end of the previous DeepShallow simulation block.

### 3.2 Emissions

Hourly emissions used in this study are the same as those used by Shrivastava et al. (2013). In brief, hourly emissions of aerosol and trace gases are derived for the desired 2007 period by assuming a linear variation in the U.S. Environmental Protection Agency's National Emissions Inventory (NEI; e.g., <http://www.epa.gov/ttn/chief/net/2005inventory.html>) for 2005 and 2008, supplemented with biomass burning gas and aerosol emissions taken from the 2007 Fire Inventory produced by NCAR (FINN07) (Wiedinmyer et al., 2011). The NEI contains two sizes of

particulate matter emissions: particles with diameters less than or equal to 2.5  $\mu\text{m}$  ( $\text{PM}_{2.5}$ ) and those less than or equal to 10  $\mu\text{m}$  ( $\text{PM}_{10}$ ). NEI  $\text{PM}_{2.5}$  emissions are divided into categories of sulfate, nitrate, organic aerosol, elemental carbon, and unspciated primary  $\text{PM}_{2.5}$ , following Hsu et al. (2006). As in Shrivastava et al. (2013), all unspciated  $\text{PM}_{2.5}$  is lumped into the MOSAIC other inorganic material (OIN) category. For the simulations presented here, OIN accounts for approximately 77% of the  $\text{PM}_{2.5}$  mass emissions. The MOZART model (Emmons et al., 2010a) was used to provide the inflow of dust through the boundaries of the WRF-Chem domain with these values assumed to be OIN.  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  emissions are mapped to eight size bins for the sectional size distribution representation following Fast et al. (2006). Particles in each size bin are assumed to be internally mixed and the same size distribution is assumed for all species. VOC emissions were speciated using the SAPRC-99 mechanism and biogenic VOC emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN <http://bai.acd.ucar.edu>) (Guenther et al., 2006). The 138 biogenic species in MEGAN are grouped into three classes for use with WRF-Chem. Primary emissions are further modified to account for semi-volatile and intermediate volatility organic compounds (S/IVOC) that are large potential anthropogenic SOA precursors and are co-emitted with primary organic aerosols (POA) (Shrivastava et al., 2008). In this study, emissions of SVOC are assumed to be twice that of POA for anthropogenic sources, while IVOC emissions are estimated to be 1.5 times the sum of SVOC and POA emissions, for a total S/IVOC emissions equal to 6.5 times POA (Hodzic et al., 2010; Tsimpidi et al., 2010; Shrivastava et al., 2011). A two-species **Volatility Basis Set (VBS)** mechanism is used here, with both POA and secondary organic aerosols (SOA) assumed to have a very low volatility (Shrivastava et al., 2011). In previous work, Shrivastava et al. (2013) showed that this 2-species VBS mechanism resulted in reasonable predictions of organic aerosols compared to measurements made during CHAPS, as described in the next section.

## **4 Data**

In this study, a subset of model results are compared to data collected during CHAPS, which was conducted during June 2007 and included the deployment of the Department

1 of Energy's Gulfstream-1 (G-1) aircraft and National Aeronautics and Space  
2 Administration (NASA) Langley Research Center B200 aircraft. During CHAPS the G-1  
3 was configured for *in situ* sampling of aerosol chemical and optical properties (Berg et  
4 al., 2009). The flight path was specifically designed to measure conditions below, within,  
5 and above a population of shallow cumuli near Oklahoma City, Oklahoma. The size  
6 distribution of aerosol and cloud drops was measured using a Droplet Measurement  
7 Technology (DMT) Passive Cavity Aerosol Spectrometer Probe (PCASP) and DMT  
8 Cloud Aerosol Spectrometer (CAS). The G-1 was equipped with two aerosol inlets: an  
9 isokinetic inlet for sampling aerosol in clear air and interstitial aerosol within clouds, and  
10 a Counter Flow Virtual Impactor (CVI) to sample only cloud droplets. An Aerodyne  
11 Aerosol Mass Spectrometer (AMS) was used to analyze the composition of non-  
12 refractory aerosol sampled via both inlets. In their work, Shrivastava et al. (2013)  
13 evaluated the performance of WRF-Chem for the same period and found reasonable  
14 agreement with the observations when the model was run with relatively fine spatial  
15 resolution (2 km) that explicitly represented convection. They reported some  
16 discrepancies between the simulated and observed aerosol optical properties, but these  
17 were attributed to assumptions related to the emissions, hygroscopicity, and complex  
18 index of refraction of of OIN particles, in addition to aerosol water content.

19 The B200 was equipped with the downward looking NASA Langley high spectral  
20 resolution lidar (HSRL-1) that provided height resolved observations of aerosol  
21 backscatter, extinction, and depolarization that were nearly simultaneous with the *in situ*  
22 G-1 measurements. Details of the HSRL-1 system can be found in Hair et al. (2008). The  
23 HSRL-1 uses the spectral distribution of the lidar return signal to separate the molecular  
24 and aerosol signal and can independently determine the aerosol backscatter, extinction,  
25 and depolarization at a wavelength of 532 nm. The HSRL-1 also functions as a standard  
26 backscatter lidar at a wavelength of 1064 nm, measuring both backscatter and  
27 depolarization at that wavelength. During CHAPS, the B200 aircraft flew above the G-1,  
28 providing lidar "curtains" along the flight track.

## 5 Analysis

In a previous case study, Berg et al. (2013) showed that the use of the KF-CuP parameterization in WRF led to a significant increase in the amount of simulated shallow sub-grid convective clouds for three days in 2007 (16 May, 2 July, and 24 July) over the Department of Energy's Atmospheric Radiation Measurement (ARM) Central Facility, consistent with observations. **In contrast, the standard KF scheme did a poor job representing the shallow clouds in these cases.** Therefore, the performance of the cumulus parameterization will not be rigorously evaluated here. A single example of the model's ability to simulate the observed cloud fields is illustrated in Figure 2 that shows the GOES visible image (valid at 20:15 UTC) and the cloud fraction associated with sub-grid clouds simulated by the cumulus parameterization and areas with grid resolved clouds at 20:00 UTC on 25 June 2007. The KF-CuP parameterization predicts large areas with shallow convection over much of the central United States, which is consistent with the areas of shallow cumuli seen in the satellite image over much of Iowa, Kansas and Missouri, and a number of deep convective clouds over Texas and Oklahoma. The frequency of occurrence in which shallow or deep convection were triggered in the WRF grid columns for the period 12:00-20:00 UTC on 25 June 2007 is shown in Figure 3 and provides information about the air-mass history in regards to sub-grid cumuli within the three boxes. Note that there can be cases in which the color shading indicates both shallow and deep clouds in the same model grid column. This occurs when different cloud types occur at different times of day.

Due to the spatial inhomogeneity of the cloud fields over the central United States highlighted in Figure 2, our analysis of conditions on 25 June will focus on three different distinct regions each approximately 240 km on a side, not just the CHAPS area around Oklahoma City that was analyzed by Shrivastava et al. (2013). These areas, approximately centered on Madison, Wisconsin (MSN); Austin, Texas (AUS); and Oklahoma City, Oklahoma (OKC), were selected because they contain primarily shallow convection (MSN), deep convection (AUS), or a mixture of both (OKC) (see Figure 3) and allow us to better understand the behavior of the model and its parameterizations over a range of conditions. The MSN box has a very high frequency of shallow clouds distributed over the box with the nearest up-stream deep convection occurring over

1 central Illinois. The AUS box has a very small frequency of simulated sub-grid shallow  
2 clouds and a much larger frequency of simulated sub-grid deep convective clouds. In  
3 contrast to the other two boxes, the OKC box includes a mixture of both shallow and  
4 deep convection.

5 While MOSAIC represents multiple aerosol constituents, only BC, OA, and sulfate have  
6 been selected for analysis within the three boxes. These particular constituents were  
7 selected because of their climatic relevance, and their representative behavior. BC is, to a  
8 first approximation, only impacted by transport, activation/resuspension, dry deposition,  
9 and wet removal—and in the case of non-precipitating convection acts essentially as a  
10 passive tracer. Although freshly emitted BC is hydrophobic, the internal mixing  
11 assumption applied in the model causes it to quickly reside in hygroscopic particles.  
12 Interpretation of cloud-aerosol interactions and vertical sulfate transport is more  
13 complicated than for BC because sulfate can be produced within cloud droplets via  
14 aqueous-phase oxidation of dissolved sulfur dioxide gas as well as removed via  
15 precipitation (e.g. Easter and Hobbs, 1974; Hegg et al., 1986; Tremblay and Leighton,  
16 1986; Chaumerliac et al., 1987; Taylor, 1989; Wang and Chang, 1993; Koch et al., 2003).  
17 While the majority of OA in the atmosphere is secondary and is somewhat hygroscopic,  
18 its behavior within convective clouds is similar to that of BC aerosol because the aqueous  
19 chemistry related to OA production is not fully understood and currently is not included  
20 in the model.

### 21 **5.1.1 Local Impacts on Aerosol Vertical Distribution**

22 One important impact of convective clouds is the vertical redistribution of aerosol due to  
23 the impact of convective updrafts, downdrafts, entrainment mixing, enhanced subsidence,  
24 and wet removal associated with sub-grid clouds. Figure 4 shows examples of vertical  
25 north-south cross sections (through the center of the analysis boxes) of the amount of BC  
26 (including both interstitial and activated aerosol in the cloudy grid cells) for the  
27 DeepShallow case and the fractional change in BC loading between the DeepShallow and  
28 control simulations (indicated by the colors) as well as the cloud fraction (indicated by  
29 the gray shading) within the AUS and MSN boxes valid at 20:00 UTC on 25 July, 2007.  
30 Within both the AUS and MSN boxes the largest BC mass loadings are found near the



1 surface. There are also large amounts of BC 4-6 km above the surface in the AUS cross  
2 section that is apparent in both the DeepShallow (Figure 4) and Control simulations (not  
3 shown). This elevated layer is not associated with convection but rather with long-range  
4 transport, most likely from a fire located in central New Mexico (not shown) and a coal-  
5 fired power plant in Colorado.

6 At first glance it might be surprising that there are not columns of enhanced aerosol  
7 loading within the AUS clouds due to enhanced upward transport from the sub-cloud  
8 layer shown in Figure 4. Their absence is primarily due to the wet removal of aerosol  
9 within the lowest levels of the clouds, as well as the cloud fraction (which ranges from 20  
10 to 60% within the deep convective clouds shown in the figure), which reduces the  
11 relative impact of the aerosol in the updraft, within any given model grid cell. In the AUS  
12 cross section, the large fractional increase in BC between the DeepShallow and Control  
13 simulations for altitudes ranging from 3 and 5 km and the decrease above 5 km can be  
14 attributed to vertical transport by updrafts, downdrafts, and convection induced  
15 subsidence. At these altitudes (which are below the detrainment level), this transport  
16 replaces some of the air (and aerosol) in a grid cell with air from higher levels that has  
17 smaller BC concentrations.

18 Within the AUS cross section, the clouds extend from an altitude of approximately 0.5  
19 km to nearly 15 km. The clouds in the MSN box are much shallower, extending from  
20 approximately 1 to 2 km as is more typical for boundary-layer cumuli (e.g. Berg and  
21 Kassianov, 2008). The decrease in amount of BC loading in the sub-cloud layer is caused  
22 by the venting of aerosol out of that layer by the convective clouds. In contrast to the  
23 AUS box that includes deep sub-grid convective clouds, the vertical extent of the  
24 transport of BC is more limited within the MSN box (Figure 4b). This result is consistent  
25 with the much smaller vertical extent of the clouds in this box. Within the cloud layer, the  
26 shallow cumuli still have an important impact on the vertical extent of the BC (Figure  
27 4d). The fractional difference in the BC between the DeepShallow and Control  
28 simulations approaches 50% as the convective clouds transport BC from below the cloud  
29 into the cloud layer. The net effect of the non-precipitating cumuli is to mix BC over the  
30 sub-cloud and cloud layers, which is consistent with the findings of others (e.g. Vilà-  
31 Guerau de Arellano et al., 2005; Kim et al., 2012).

1 Similar to the case for BC, there is an elevated plume of sulfate aerosol near an altitude of  
2 5 km in the AUS cross-section that is associated with long-range transport (Figure 5a). In  
3 both the AUS and MSN cross-sections there is a large concentration of sulfate within the  
4 boundary layer that is associated with surface emissions. As with BC, fractional  
5 differences between the DeepShallow and Control runs are much larger than 50%. Within  
6 the AUS box there is a large fractional change in the amount of sulfate aloft that can be  
7 attributed to vertical transport by updrafts, downdrafts, and convection induced  
8 subsidence that are represented in the DeepShallow simulations (Figure 5c). The situation  
9 is different in the MSN box, where all of the clouds are shallow non-precipitating cumuli  
10 (Figure 5b and d). In this case, the vertical transport is limited to the cloud layer (altitudes  
11 lower than approximately 2 km), where there is significant increase in the sulfate loading  
12 in the cloud layer (Figure 5b). In contrast to the BC within the MSN box, the sulfate is  
13 enhanced in the ShallowOnly simulations both below and within the cloud layer. This is  
14 due to sulfate production within clouds, the detrainment of cloudy air with enhanced  
15 sulfate, and subsequent downward transport of air back into the subcloud layer. There is  
16 no evidence of lofted sulfate in the levels above the shallow cumuli (Figure 5b).

17 Using data from the G-1 aircraft alone, it is difficult to verify the simulation of the  
18 vertical transport of aerosol associated with cumulus. Data from the airborne NASA  
19 HSRL, however, can be used to investigate the vertical extent of aerosol in the vicinity of  
20 convective clouds. This data set does not provide information in regards to the speciation  
21 of the aerosol, but it can be used to look at impacts on the aerosol backscatter and  
22 extinction, which are highly correlated with the aerosol loading. Unfortunately, HSRL  
23 data are not available for 25 June, so two other days have been selected for analysis of  
24 vertical transport, including 19 and 21 June 2007. The frequency of simulated shallow  
25 and deep clouds are shown in Figure 6, and both days had relatively large amounts of  
26 simulated shallow clouds both before and during the G-1 and B200 flights.

27 Conditions on 19 June were marked by large amounts of both observed and simulated  
28 shallow cumuli near Oklahoma City and are similar to the MSN grid box on 25 June. In  
29 most cases the observed shallow cumuli are sufficiently optically thick that the laser  
30 beam is attenuated by the cloud, leading to the frequent periods of missing data below  
31 cloud top (as indicated by the white areas underneath peaks in the aerosol backscatter in

Figure 7). On 19 June the majority of cloud-top heights measured by the HSRL are found to range from 1 to 2.5 km and there are relatively large amounts of aerosol backscatter and extinction from the surface to an altitude of 2.5 km, which roughly corresponds to the highest cloud top heights observed during the B200 flight. The DeepShallow and Control simulations were subsampled along the B200 flight track and the fractional difference in the WRF-Chem simulated extinction and cloud fraction associated with convective clouds is shown in Figure 7c. Both the DeepShallow and Control simulations underestimate the aerosol extinction on 19 June by approximately a factor of 1.25 to 2.0 compared to the values derived from the HSRL (not shown). This is likely due to both an underestimate of aerosol mass loading as well as an underestimate of the simulated water uptake by the aerosol. Given that both sets of simulations underestimate the observed values, the underestimate of backscatter and extinction is not attributable to the treatment of sub-grid convective clouds. The simulated cloud fraction reaches values as large as 40% and the vertical extent of the simulated clouds is consistent with the HSRL observations. The largest positive differences in the simulated extinction are associated with the layer of shallow cumuli. The enhanced transport associated with the DeepShallow simulations spread the aerosol, and hence aerosol extinction, over a layer from the surface to an altitude of 2 km, compared to only 1 km in the control simulations. The values of extinction in the DeepShallow simulations are 10-20% greater over altitudes ranging from 1 to 2 km than was found in the Control simulations, while the extinction in the subcloud layer is reduced by a similar magnitude. This behavior is similar to the changes in the seen with the BC loading within the MSN analysis box on 25 June (Figure 4). The differences between the DeepShallow and ShallowOnly simulations are subtle on 19 June (not shown), because of the relatively small amount of deep convection in the vicinity of the OKC analysis box. There is, however, still a decrease in the aerosol mass loading in the DeepShallow simulations compared to the ShallowOnly case.

In contrast to 19 June, which had a large fraction of shallow convection and very few deep clouds near Oklahoma City, conditions on 21 June were marked by a mixture of deep and shallow clouds in both the observations and simulations. The HSRL data shows a region of higher clouds (near 20:00 UTC; Figure 8). The tops of the observed shallow

clouds range from approximately 1 to 2 km and are distributed along much of the entire flight track. The vertical transport associated with the clouds leads to enhanced aerosol backscatter and extinction to an altitude of 2 km. There are fewer simulated clouds along the flight track at the time of the B200 flights than is observed, and there are some simulated deep convective clouds between 19:30 and 20:00 UTC and near 21:15 UTC (Figure 8). There are some systematic changes in the simulated aerosol extinction, suggesting additional clouds upwind of the flight track, or clouds that occur before the B200 was aloft. These changes include an increase in the aerosol extinction in the DeepShallow simulations near an altitude of 1.75 km between 19:00 to nearly 20:00 UTC, and near an altitude of 2 km from 20:15 through approximately 21:30 UTC. The results for 21 June are reminiscent of changes seen in the AUS analysis box for 25 June and suggest an increased impact of deep convection on 21 June than was seen on 19 June.

There are differences between the results of the DeepShallow simulation shown in Figure 8 and the ShallowOnly simulation (not shown) that demonstrate the relative importance of the deep clouds. The ShallowOnly simulation has more extinction for altitudes larger than 1.5 km, which is associated with less wet removal and less vertical transport. This is particularly the case around 20:30 UTC, where the DeepShallow simulation has a large decrease in extinction associated with the deep clouds (Figure 8). While there are no deep convective clouds shown in the lidar cross section (there were some clouds with tops as high as 5 km), it is difficult to evaluate the relative skill of the parameterization to predict the occurrence of deep convective clouds from a single cross section. It should be noted that on this particular day there were number of both simulated and observed deep clouds near the sampling domain.

## 5.2 Regional scale impacts

The results presented in Section 5.1 highlight that the parameterization is performing reasonably and can be used to investigate the regional impacts of cloud-aerosol interactions within the areas defined by the analysis boxes. The primary advantage of using a parameterization to represent convective clouds is the ability to run simulations over a large domain, which enables the evaluation of regional scale impacts of cloud-aerosol interactions on the aerosol lifecycle that is not possible using high-resolution

1 simulations. Differences in the column-integrated mass loading are one method that can  
2 be used to investigate changes in mass loading of atmospheric aerosol over large areas.  
3 BC represents particles that are essentially passive tracers (ignoring wet and dry removal)  
4 that do not undergo aqueous phase chemistry in simulated clouds. Overall, there is a  
5 significant reduction in the column integrated BC and OA across the model domain  
6 (Figure 9). The primary removal mechanism added in the DeepShallow simulations  
7 (compared to the Control simulations) is the wet removal associated with the  
8 parameterized precipitation. This leads to systematic decreases of as much as -50% in the  
9 amount of BC. It is interesting to note that there is a net decrease of BC within the MSN  
10 box in which there is no convection and very little grid resolved precipitation, indicative  
11 of wet removal upwind of the box during the simulation and pointing to regional scale  
12 impacts of cloud-aerosol interactions. There are also small areas in which the column  
13 integrated BC loading is larger in the DeepShallow than control simulations. These  
14 features are also present in the ShallowOnly case (not shown). The increase in the  
15 column integrated BC in the AUS box is the result of slight differences in the path of the  
16 aerosol plume coming from the Houston, Texas area. Different aerosol loadings in the  
17 simulations produce different feedbacks on meteorology (i.e., aerosol indirect effects in  
18 grid-resolved clouds and aerosol direct effects), leading to small differences in winds. In  
19 the DeepShallow simulations the main part of the plume is shifted a small distance to the  
20 north, giving rise to the apparent increase in the BC loading just downwind of Houston.

21 The OA follows a pattern similar to what is seen for BC, but the fractional change is  
22 smaller in magnitude. Currently in WRF-Chem the OA are unaffected by aqueous  
23 chemistry within the clouds, but can be affected by changes in the amount of precursor  
24 gases. Vertical transport of SOA precursor gases (which are not wet-removed in our  
25 parameterization) to higher and colder altitudes can result in more partitioning to the  
26 particle phase. These changes lead to areas, such as the central swath through the OKC  
27 box, and over parts of the southeastern United States, where there is an increase in the  
28 column integrated OA. Based on these simulations the change in OA can be significant,  
29 approaching a column integrated increase of 10 to 15% for some areas.

1 In contrast to BC, the wet removal of sulfate can be counteracted by its production in  
2 cloud. In the AUS box, sulfate wet removal is larger than production, leading to a small  
3 net decrease in sulfate when cloud-aerosol interactions (including aqueous chemistry)  
4 associated with deep and shallow clouds are considered. Within the MSN box (and over  
5 much of the upper-midwest), there is no convective and very little grid resolved  
6 precipitation so that the production of sulfate aerosol by aqueous chemistry dominates  
7 and there is a significant increase in the column burden of sulfate when non-precipitating  
8 clouds are present (Figure 9). The additional sulfate is limited to the cloud layer and  
9 below, but as shown in Figure 5 this enhanced sulfate can spread over a deeper layer of  
10 the atmosphere. Relative to the control case where the impacts of cumulus are ignored,  
11 our results indicate that cumulus can increase the column sulfate burden by as much as  
12 40%. While the simulations shown here were rather short, longer integration times could  
13 lead to significant differences downwind of the area of sulfate production due to  
14 enhanced vertical mixing and regional scale transport.

### 15 **5.3 Impact on cloud microphysics**

16 Using data collected during CHAPS, Berg et al. (2011) measured differences in cloud  
17 microphysical properties that could be explained by differences in the aerosol  
18 concentrations and vertical velocities within individual clouds . They used the carbon  
19 monoxide mixing ratio perturbation ( $CO'$ ; defined as the difference between the  
20 instantaneous measured CO and the average CO observed during a flight leg) as an  
21 indicator of increased aerosol. They found systematic increases in the cloud droplet  
22 number concentration (CDNC) associated with both increases in  $CO'$  and the cloud  
23 updraft strength, which highlighted the importance of considering both the aerosol  
24 loading and the cloud dynamics. In their analysis of high-resolution WRF-Chem  
25 simulations, Shrivastava et al. (2013) found results consistent with those reported by Berg  
26 et al. (2011). A similar analysis has been completed here using results from the  
27 DeepShallow simulations in the OKC analysis box, but limited to only grid columns with  
28 shallow convection. The cloud microphysical properties were computed for only the  
29 cloudy updrafts, as this is the part of the parameterized clouds where the sub-cloud  
30 particle loading can influence the cloud microphysical properties via drop activation. A

1 probability density function (PDF) of simulated CO' and perturbation vertical velocity  
2 ( $w'$ , defined in a way analogous to CO') is shown in Figure 10. In this case the  
3 parameterized updraft speeds were found to range from 1.0 to 3.5 m s<sup>-1</sup> which are  
4 consistent with the updraft speeds in Figure 1 of Berg et al. (2011).

5 For the parameterized sub-grid convective clouds the CDNC is found to increase with  
6 increasing values of CO, showing an increase from about 500 to 800 cm<sup>-3</sup> (an increase by  
7 about a factor of 1.6) as the CO' ranges from clean (-35 ppbv) to dirty (+35 ppbv) for  
8 model grid cells where the updraft ranges from 2.0 to 2.5 ms<sup>-1</sup> (Figure 10). The results are  
9 fairly noisy with relatively large standard deviations highlighting the wide range of  
10 additional factors that can impact the CDNC. The slope of the CDNC vs. CO' regression  
11 line for  $w'$  equal to 2.0-2.5 m s<sup>-1</sup> is computed to be 4.2 cm<sup>-3</sup> ppbv<sup>-1</sup>, which is smaller than  
12 the 7.2 cm<sup>-3</sup> ppbv<sup>-1</sup> reported by Berg et al. (2011), but is close to the value of 4.5 cm<sup>-3</sup>  
13 ppbv<sup>-1</sup> derived from the results of Shrivastava et al. (2013). The different slopes seen in  
14 the observations, those reported by Shrivastava et al. (2013) and this study could be  
15 related to the smoothing of emissions, which has been documented in the literature in  
16 regards to both simulated cloud characteristics (Gustafson et al., 2007) and aerosol  
17 loadings (Gustafson et al., 2011). The results shown by the different studies should be  
18 considered with care, however, because of the different vertical velocity ranges used in  
19 each case. While not ideal, the different  $w$  ranges were applied because of differences in  
20 the spatial and temporal scales associated with the observations and high- and low-  
21 resolution simulations.

## 22 **5.4 Chemical composition of cloud drops**

23 Changes to chemical properties of the particles associated with passage through clouds  
24 are an important aspect of cloud aerosol interactions. One of the goals of the CHAPS  
25 study was to document changes in the chemical composition of particles that served as  
26 CCN (activated) or remained inactive (interstitial). During CHAPS, measurements  
27 showed both the activated and interstitial aerosol were dominated by organics and sulfate  
28 (Figure 11). In their analysis, Berg et al. (2009) also reported enhanced nitrate in the  
29 dried cloud drop residuals that were sampled via a counter flow virtual impactor (CVI),  
30 consistent with model results presented by Hegg et al. (1986). They attributed this to the

1 uptake of gas-phase nitric acid by cloud drops. In their analysis of high-resolution WRF-  
2 Chem simulations, Shrivastava et al. (2013) also found enhanced nitrate when aqueous  
3 phase chemistry, which includes trace gas-liquid phase equilibria, was turned on. When  
4 aqueous phase chemistry was turned off **in their simulations**, however, the particle nitrate  
5 in cloud drop residuals and interstitial particles was nearly the same, indicating the  
6 importance of the uptake and dissociation of gas-phase nitric acid within cloud drops.

7 A similar analysis has been completed for the OKC box using results from ShallowOnly  
8 simulations. The mass loading of the interstitial aerosol within the shallow clouds is  
9 generally smaller in this study than the loading reported by Shrivastava et al. (2013) for  
10 either the observations (Figure 11) or high-resolution simulations (Figure 7 of  
11 Shrivastava et al., 2013). This behavior may, in part, be attributed to the averaging of the  
12 emissions over the larger model grid cell in the vicinity of Oklahoma City and the  
13 location of the simulated shallow clouds in the two studies. In contrast to the interstitial  
14 particles, the simulated mass loading of the activated aerosol is larger in all three  
15 simulations (grid-resolved, ShallowOnly with cloud chemistry on, and ShallowOnly with  
16 cloud chemistry off) than the loading that was observed during CHAPS. The over-  
17 estimation of simulated aerosol mass may, in part, be due to the cut size used by the CVI  
18 operated on the aircraft that would exclude small cloud drops. In contrast to the aerosol  
19 mass loading, the observed and simulated aerosol volume fractions are in good  
20 agreement. Thus, even if the mass loading is incorrect, the consistent volume fractions  
21 indicates that the chemical processing within the model clouds is behaving in a way that  
22 is consistent with the observations. Similar to the observations and high-resolution  
23 simulations, there is an increase in the volume fraction of nitrate in activated (cloud-  
24 borne) aerosol compared to interstitial aerosol.

25 The analysis of activated versus interstitial aerosol composition is repeated for the  
26 CHAPS flights on 20 and 23 June. These days also had shallow cumuli in the vicinity of  
27 Oklahoma City, although the simulated cloud fraction (not shown) was less than  
28 **observed** on 19, 21, and 25 June. On 20 June, the median organic volume fractions of  
29 interstitial aerosol were approximately 75% organics and 15% sulfate, with small  
30 amounts of nitrate and ammonium (Figure 12a). The activated aerosol sampled using the  
31 CVI were also dominated by organics, but there was a great deal of variability in the



1 volume fraction of organics as well as an increase in the volume fraction of nitrate. The  
2 ShallowOnly simulations for the OKC box for 20 June are consistent with the observed  
3 values and follow the same trends for both interstitial and activated aerosol. The  
4 variability in the simulated volume fraction is much less than was observed, which could  
5 be a result of the relative small amount of simulated sub-grid convective clouds on that  
6 day. The simulated activated aerosol also had enhanced values of nitrate aerosol  
7 compared to the interstitial aerosol. Observations on 23 June include a smaller volume  
8 fraction (60%) of organics in the case of interstitial aerosol, and increased volume  
9 fraction of sulfate (30%) compared to the other two days (Figure 11 and Figure 12b). The  
10 volume fraction of activated aerosol is also dominated by organics, but like conditions on  
11 20 June, there is a great deal of variability. The ShallowOnly simulations have a  
12 relatively large median volume fraction associated with sulfate aerosol that is consistent  
13 with observations and smaller amounts of organic aerosol than was seen on 20 and 25  
14 June. The simulations also have enhanced nitrate volume fraction compared to the  
15 interstitial aerosol. Thus the increase in nitrate aerosol seen in both the observations and  
16 simulations associated with aqueous chemistry is not limited to a single day, but rather is  
17 found to be a relatively common occurrence in the OKC box during CHAPS.

## 18 **Summary and Conclusions**

19 A new treatment of cloud-aerosol interactions within parameterized shallow and deep  
20 convection has been implemented in WRF-Chem with the goal of improving regional  
21 scale simulations of the aerosol lifecycle and cloud-aerosol interactions. The  
22 modifications designed to represent cloud-aerosol interactions include treatment of the  
23 cloud droplet number mixing ratio; key cloud microphysical and macrophysical  
24 parameters (including the updraft fractional area, updraft and downdraft mass fluxes, and  
25 entrainment) averaged over the population of shallow clouds, or a single deep convective  
26 cloud; and vertical transport, activation/resuspension, aqueous chemistry, and wet  
27 removal of aerosol and trace gases in warm clouds. These changes have been  
28 implemented in the WRF-Chem chemistry package as well as the Kain-Fritsch cumulus  
29 parameterization (Kain and Fritsch, 1990; Kain, 2004), which has been modified to better  
30 represent shallow convective clouds (Berg et al., 2013). Results from simulations using

1 WRF-Chem are compared with data from the CHAPS field experiment (Berg et al., 2009;  
2 Berg et al., 2011) as well as high-resolution simulations (Shrivastava et al., 2013).

3 The results are encouraging and demonstrate the advantages of the modifications that  
4 have been made to WRF-Chem. It is shown that both deep and shallow convective clouds  
5 have an important impact on the horizontal and vertical distribution of aerosol loading.  
6 Three different domain sub-regions were selected for detailed analysis, including  
7 locations near Madison, Wisconsin (MSN), Austin, Texas (AUS), and Oklahoma City,  
8 Oklahoma (OKC), the **last** corresponding to the site of CHAPS and the domain used in  
9 previous high-resolution simulations. These regions were selected to represent instances  
10 dominated by shallow (MSN), deep (AUS), or a mix of both (OKC) types of convective  
11 clouds. In each case the WRF-Chem simulations behaved in a manner consistent with  
12 expectations and consistent with both the CHAPS data and the results of high-resolution  
13 simulations. In the case of shallow clouds, enhanced mixing leads to a deepening of the  
14 layer containing BC and decreased amounts of BC near the surface. Results are similar  
15 for OA, but the net impact was found to be smaller. In contrast to BC, sulfate aerosol was  
16 enhanced throughout the layer due to sulfate production within clouds. In the vicinity of  
17 AUS, the impact of shallow convective clouds is minimal. There was a decrease in BC,  
18 OA, and sulfate in the sub-cloud layer due to vertical transport associated with deep  
19 convective clouds. There were also significant changes in the aerosol loading aloft that  
20 were the result of the impacts of updrafts, downdrafts, entrainment mixing, enhanced  
21 subsidence, and wet removal associated with the sub-grid clouds. In the area near OKC,  
22 both the deep and shallow sub-grid convective clouds had a significant impact on the  
23 simulated aerosol loading. The shallow sub-grid clouds led to a decrease of aerosol in the  
24 sub-cloud layer and an increase of aerosol aloft. The parameterized deep-convective  
25 clouds led to decreases in the BC and OA over the lowest 2 km and sulfate over the  
26 lowest 3 km of the atmosphere.

27 One of the motivations for the development of the improved parameterization is to allow  
28 the investigation of regional and synoptic scale aerosol transport. In our case-study  
29 period, there is a significant reduction in the BC and OA over much of the central United  
30 States. The primary removal mechanism added in the new treatment is the wet removal  
31 associated with the parameterized precipitation. Thus, the differences in the aerosol

loading highlight the importance of wet removal on the aerosol lifecycle at the regional scale. In contrast to BC and OA, there are large regions in which there are increases in the column-integrated sulfate due to the production of sulfate and absence of wet removal in nonprecipitating clouds.

The behavior of the modified version of WRF-Chem in regards to the cloud microphysical properties and chemical composition of aerosol is also investigated. The results show that the modified version of WRF-Chem is able to reproduce changes in the cloud droplet number concentration in a way that is consistent with both high-resolution simulations and observations from CHAPS. The CDNC associated with the parameterized clouds was found to be less sensitive to pollutant loading than was observed (Berg et al., 2011) but was similar to that reported by Shrivastava et al. (2013) in their high resolution simulations. The chemical composition of the simulated cloud-drop residuals is compared to the composition measured with an AMS operated behind a CVI inlet during CHAPS. While there were differences in the simulated and observed mass loadings, the simulated and observed mass fractions were consistent, including the presence of enhanced amounts of nitrate in the cloud drop residuals. WRF-Chem is also able to accurately represent the increase in nitrate found in the observed cloud-drop residuals. Overall, these findings provide evidence that the modified version of WRF-Chem is able to represent key features of the cloud-aerosol interactions in a realistic way. While the results presented here utilized WRF-Chem version 3.2.1, the code is being ported to the latest version of WRF-Chem and we anticipate including these changes in a future public release of WRF-Chem.

## **Acknowledgements**

This research was supported by the Office of Science of the U.S. Department of Energy as part of the Atmospheric System Research Program (ASR). The Pacific Northwest National Laboratory is operated by Battelle Memorial Institute under contract DE-AC06-76RLO 1830. The funding for the B200 and HSRL operations came from the NASA Science Mission Directorate, the Department of Energy ASR program, and the NASA CALIPSO project. The authors would also like to thank the NASA Langley King Air B-

1 200 and DOE G-1 flight crews for their outstanding work supporting these flights and  
2 measurements. Drs. J. Ogren of NOAA and E. Andrews of the Cooperative Institute for  
3 Research in Environmental Sciences (CIRES) deployed the CVI during CHAPS. Data  
4 from the AMS was collected by Drs. Y.-N. Lee of Brookhaven National Laboratory  
5 (BNL), M. L. Alexander of Pacific Northwest National Laboratory and J. Jayne of  
6 Aerodyne. Size distribution data were provided by Dr. G. Senum of BNL. Drs. G. Grell  
7 (NOAA) and R. Leung (PNNL) provided feedback on various aspects of the manuscript.  
8 We also thank **three anonymous reviewers valuable feedback on the manuscript.**  
9

1

## 2    **6    References**

3    Abdul-Razzak, H., and Ghan, S. J.: A parameterization of aerosol activation 2. Multiple  
4    aerosol types, *J. Geophys. Res.*, 105, 6837-6844, 10.1029/1999jd901161, 2000.

5    Ahmadov, R., McKeen, S. A., Robinson, A. L., Bahreini, R., Middlebrook, A. M., de  
6    Gouw, J. A., Meagher, J., Hsie, E. Y., Edgerton, E., Shaw, S., and Trainer, M.: A  
7    volatility basis set model for summertime secondary organic aerosols over the eastern  
8    United States in 2006, *J. Geophys. Res.*, 117, D06301, 10.1029/2011jd016831, 2012.

9    Alapaty, K., Herwehe, J. A., Otte, T. L., Nolte, C. G., Bullock, O. R., Mallard, M. S.,  
10    Kain, J. S., and Dudhia, J.: Introducing subgrid-scale cloud feedbacks to radiation for  
11    regional meteorological and climate modeling, *Geophys. Res. Lett.*, 39, L24809,  
12    10.1029/2012gl054031, 2012.

13    Arakawa, A., Jung, J. H., and Wu, C. M.: Toward unification of the multiscale modeling  
14    of the atmosphere, *Atmos. Chem. Phys.*, 11, 3731-3742, 10.5194/acp-11-3731-2011,  
15    2011.

16    Barahona, D., and Nenes, A.: Parameterization of cloud droplet formation in large-scale  
17    models: Including effects of entrainment, *J. Geophys. Res.*, 112, 10.1029/2007jd008473,  
18    2007.

19    Berg, L. K., and Stull, R. B.: Parameterization of Joint Frequency Distributions of  
20    Potential Temperature and Water Vapor Mixing Ratio in the Daytime Convective  
21    Boundary Layer, *J. Atmos. Sci.*, 61, 813-828, 10.1175/1520-  
22    0469(2004)061<0813:pojfd>2.0.co;2, 2004.

23    Berg, L. K., and Stull, R. B.: A Simple Parameterization Coupling the Convective  
24    Daytime Boundary Layer and Fair-Weather Cumuli, *J. Atmos. Sci.*, 62, 1976-1988,  
25    10.1175/jas3437.1, 2005.

26    Berg, L. K., and Kassianov, E. I.: Temporal Variability of Fair-Weather Cumulus  
27    Statistics at the ACRF SGP Site, *J. Climate*, 21, 3344-3358, 10.1175/2007jcli2266.1,  
28    2008.

29    Berg, L. K., Berkowitz, C. M., Hubbe, J. M., Ogren, J. A., Hostetler, C. A., Ferrare, R.  
30    A., Hair, J. W., Dubey, M. K., Mazzoleni, C., Andrews, E., Coulter, R. L., Lee, Y.-N.,  
31    Olfert, J., and Springston, S. R.: Overview of the Cumulus Humilis Aerosol Processing  
32    Study, *Bull. Amer. Meteor. Soc.*, 90, 1653-1667, 10.1175/2009bams2760.1, 2009.

33    Berg, L. K., Berkowitz, C. M., Barnard, J. C., Senum, G., and Springston, S. R.:  
34    Observations of the first aerosol indirect effect in shallow cumuli, *Geophys. Res. Lett.*,  
35    38, 10.1029/2010gl046047, 2011.

1 Berg, L. K., Gustafson, W. I., Kassianov, E. I., and Deng, L.: Evaluation of a Modified  
2 Scheme for Shallow Convection: Implementation of CuP and Case Studies, *Mon. Wea.*  
3 *Rev.*, 141, 134-147, 10.1175/mwr-d-12-00136.1, 2013.

4 Boucher, O., Randall, D. A., Artaxo, P., Bretherton, C. S., Feingold, G., Forster, P.,  
5 Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K.,  
6 Sherwood, S., Stevens, B., and Zhang, X. Y.: Clouds and Aerosols, in: *Climate Change*  
7 *2013: The Physical Science Basis. Contribution of Working Group I to the Fifth*  
8 *Assessment Report to the Intergovernmental Panel on Climate Change*, edited by: Stocker,  
9 T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y.,  
10 Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, United Kingdom  
11 and New York, NY, USA, 2013.

12 Carter, W. P. L.: SAPRC-99 Mechanism Files and Associated Programs and Examples:  
13 <http://www.cert.ucr.edu/~carter/SAPRC99/>, last updated March 30, 2010, 2010.

14 Chapman, E. G., Gustafson Jr, W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M.  
15 S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model:  
16 Investigating the radiative impact of elevated point sources, *Atmos. Chem. Phys.*, 9, 945-  
17 964, 10.5194/acp-9-945-2009, 2009.

18 Chaumerliac, N., Richard, E., Pinty, J. P., and Nickerson, E. C.: Sulfur scavenging in a  
19 mesoscale model with quasi-spectral microphysics: Two-dimensional results for  
20 continental and maritime clouds, *J. Geophys. Res.*, 92, 3114-3126,  
21 10.1029/JD092iD03p03114, 1987.

22 Chen, F., Mitchell, K., Schaake, J., Xue, Y., Pan, H.-L., Koren, V., Duan, Q. Y., Ek, M.,  
23 and Betts, A.: Modeling of land surface evaporation by four schemes and comparison  
24 with FIFE observations, *J. Geophys. Res.*, 101, 7251-7268, 10.1029/95jd02165, 1996.

25 Collins, W. D., Rasch, P. J., Boville, B. A., Hack, J. J., McCaa, J. R., Williamson, D. L.,  
26 Kiehl, J. T., Briegleb, B., Bitz, C., Lin, S.-J., Zhang, M., and Dai, Y.: Description of the  
27 NCAR Community Atmosphere Model (CAM 3.0), 2004.

28 Easter, R. C., and Hobbs, P. V.: The Formation of Sulfates and the Enhancement of  
29 Cloud Condensation Nuclei in Clouds, *J. Atmos. Sci.*, 31, 1586-1594, 10.1175/1520-  
30 0469(1974)031<1586:tfosat>2.0.co;2, 1974.

31 Eidhammer, T., Barth, M. C., Petters, M. D., Wiedinmyer, C., and Prenni, A. J.: Aerosol  
32 microphysical impact on summertime convective precipitation in the Rocky Mountain  
33 region, *J. Geophys. Res.*, 119, 2014JD021883, 10.1002/2014jd021883, 2014.

34 Emmons, L. K., Apel, E. C., Lamarque, J. F., Hess, P. G., Avery, M., Blake, D., Brune,  
35 W., Campos, T., Crawford, J., DeCarlo, P. F., Hall, S., Heikes, B., Holloway, J., Jimenez,  
36 J. L., Knapp, D. J., Kok, G., Mena-Carrasco, M., Olson, J., O'Sullivan, D., Sachse, G.,  
37 Walega, J., Weibring, P., Weinheimer, A., and Wiedinmyer, C.: Impact of Mexico City  
38 emissions on regional air quality from MOZART-4 simulations, *Atmos. Chem. Phys.*, 10,  
39 6195-6212, 10.5194/acp-10-6195-2010, 2010a.

1 Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J. F., Pfister, G. G., Fillmore, D.,  
2 Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G.,  
3 Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the  
4 Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model*  
5 *Dev.*, 3, 43-67, 10.5194/gmd-3-43-2010, 2010b.

6 Fahey, K. M., and Pandis, S. N.: Optimizing model performance: variable size resolution  
7 in cloud chemistry modeling, *Atmos. Environ.*, 35, 4471-4478,  
8 [http://dx.doi.org/10.1016/S1352-2310\(01\)00224-2](http://dx.doi.org/10.1016/S1352-2310(01)00224-2), 2001.

9 Fan, J., Leung, L. R., Li, Z., Morrison, H., Chen, H., Zhou, Y., Qian, Y., and Wang, Y.:  
10 Aerosol impacts on clouds and precipitation in eastern China: Results from bin and bulk  
11 microphysics, *J. Geophys. Res.*, 117, n/a-n/a, 10.1029/2011jd016537, 2012.

12 Fast, J. D., Gustafson, W. I., Jr., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E.  
13 G., Grell, G. A., and Peckham, S. E.: Evolution of ozone, particulates, and aerosol direct  
14 radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-  
15 aerosol model, *J. Geophys. Res.*, 111, D21305, 10.1029/2005jd006721, 2006.

16 Gerard, L., Piriou, J.-M., Brožková, R., Geleyn, J.-F., and Banciu, D.: Cloud and  
17 Precipitation Parameterization in a Meso-Gamma-Scale Operational Weather Prediction  
18 Model, *Mon. Wea. Rev.*, 137, 3960-3977, 10.1175/2009mwr2750.1, 2009.

19 Ghan, S. J., and Easter, R. C.: Impact of cloud-borne aerosol representation on aerosol  
20 direct and indirect effects, *Atmos. Chem. Phys.*, 6, 4163-4174, 10.5194/acp-6-4163-2006,  
21 2006.

22 Grell, G., Freitas, S. R., Stuefer, M., and Fast, J.: Inclusion of biomass burning in WRF-  
23 Chem: impact of wildfires on weather forecasts, *Atmos. Chem. Phys.*, 11, 5289-5303,  
24 10.5194/acp-11-5289-2011, 2011.

25 Grell, G. A.: Prognostic Evaluation of Assumptions Used by Cumulus Parameterizations,  
26 *Mon. Wea. Rev.*, 121, 764-787, 10.1175/1520-0493(1993)121<0764:peoaub>2.0.co;2,  
27 1993.

28 Grell, G. A., and Dévényi, D.: A generalized approach to parameterizing convection  
29 combining ensemble and data assimilation techniques, *Geophys. Res. Lett.*, 29, 38-31-38-  
30 34, 10.1029/2002gl015311, 2002.

31 Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C.,  
32 and Eder, B.: Fully coupled "online" chemistry within the WRF model, *Atmos. Environ.*,  
33 39, 6957-6975, 10.1016/j.atmosenv.2005.04.027, 2005.

34 Grell, G. A., and Freitas, S. R.: A scale and aerosol aware stochastic convective  
35 parameterization for weather and air quality modeling, *Atmos. Chem. Phys.*, 14, 5233-  
36 5250, 10.5194/acp-14-5233-2014, 2014.

1 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.:  
2 Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of  
3 Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181-3210, 10.5194/acp-6-  
4 3181-2006, 2006.

5 Gustafson, W. I., Qian, Y., and Fast, J. D.: Downscaling aerosols and the impact of  
6 neglected subgrid processes on direct aerosol radiative forcing for a representative global  
7 climate model grid spacing, *J. Geophys. Res.*, 116, D13303, 10.1029/2010jd015480,  
8 2011.

9 Gustafson, W. I., Ma, P.-L., Xiao, H., Singh, B., Rasch, P. J., and Fast, J. D.: The  
10 Separate Physics and Dynamics Experiment (SPADE) framework for determining  
11 resolution awareness: A case study of microphysics, *J. Geophys. Res.*, 118, 9258-9276,  
12 10.1002/jgrd.50711, 2013.

13 Gustafson, W. I., Jr., Chapman, E. G., Ghan, S. J., Easter, R. C., and Fast, J. D.: Impact  
14 on modeled cloud characteristics due to simplified treatment of uniform cloud  
15 condensation nuclei during NEAQS 2004, *Geophys. Res. Lett.*, 34, L19809,  
16 10.1029/2007gl030021, 2007.

17 Hair, J. W., Hostetler, C. A., Cook, A. L., Harper, D. B., Ferrare, R. A., Mack, T. L.,  
18 Welch, W., Izquierdo, L. R., and Hovis, F. E.: Airborne High Spectral Resolution Lidar  
19 for profiling aerosol optical properties, *Appl. Opt.*, 47, 6734-6752, 2008.

20 Hegg, D. A., Rutledge, S. A., and Hobbs, P. V.: A numerical model for sulfur and  
21 nitrogen scavenging in narrow cold-frontal rainbands: 2. Discussion of chemical fields, *J.*  
22 *Geophys. Res.*, 91, 14403-14416, 10.1029/JD091iD13p14403, 1986.

23 Herwehe, J. A., Alapaty, K., Spero, T. L., and Nolte, C. G.: Increasing the credibility of  
24 regional climate simulations by introducing subgrid-scale cloud-radiation interactions, *J.*  
25 *Geophys. Res.*, 119, 2014JD021504, 10.1002/2014jd021504, 2014.

26 Hodzic, A., Jimenez, J. L., Madronich, S., Canagaratna, M. R., DeCarlo, P. F., Kleinman,  
27 L., and Fast, J.: Modeling organic aerosols in a megacity: potential contribution of semi-  
28 volatile and intermediate volatility primary organic compounds to secondary organic  
29 aerosol formation, *Atmos. Chem. Phys.*, 10, 5491-5514, 10.5194/acp-10-5491-2010,  
30 2010.

31 Hsu, Y., Strait, R., Roe, S., and Holoman, D.: SPECIATE 4.0: Speciation Database  
32 Development Documentation 2006.

33 Janjić, Z. I.: The Step-Mountain Coordinate: Physical Package, *Mon. Wea. Rev.*, 118,  
34 1429-1443, 10.1175/1520-0493(1990)118<1429:tsmcpp>2.0.co;2, 1990.

35 Janjić, Z. I.: Nonsingular Implementation of the Mellor-Yamada Level 2.5 Scheme in the  
36 NCEP Meso model, 61, 2002.



1 Kain, J. S., and Fritsch, J. M.: A One-Dimensional Entraining/Detraining Plume Model  
2 and Its Application in Convective Parameterization, *J. Atmos. Sci.*, 47, 2784-2802,  
3 10.1175/1520-0469(1990)047<2784:aodepm>2.0.co;2, 1990.

4 Kain, J. S.: The Kain–Fritsch Convective Parameterization: An Update, *J. Applied*  
5 *Meteor.*, 43, 170-181, 10.1175/1520-0450(2004)043<0170:tkcpau>2.0.co;2, 2004.

6 Kim, S. W., Barth, M. C., and Trainer, M.: Influence of fair-weather cumulus clouds on  
7 isoprene chemistry, *J. Geophys. Res.*, 117, D10302, 10.1029/2011jd017099, 2012.

8 Koch, D., Park, J., and Del Genio, A.: Clouds and sulfate are anticorrelated: A new  
9 diagnostic for global sulfur models, *J. Geophys. Res.*, 108, 4781, 10.1029/2003jd003621,  
10 2003.

11 Larson, V. E., Schanen, D. P., Wang, M. H., Ovchinnikov, M., and Ghan, S.: PDF  
12 Parameterization of Boundary Layer Clouds in Models with Horizontal Grid Spacings  
13 from 2 to 16 km, *Mon. Wea. Rev.*, 140, 285-306, 10.1175/mwr-d-10-05059.1, 2012.

14 Lim, K.-S. S., Fan, J., Ruby Leung, L., Ma, P.-L., Singh, B., Zhao, C., Zhang, Y., Zhang,  
15 G., and Song, X.: Investigation of aerosol indirect effects using a cumulus microphysics  
16 parameterization in a regional climate model, *J. Geophys. Res.*, 2013JD020958,  
17 10.1002/2013jd020958, 2013.

18 Ma, P. L., Rasch, P. J., Fast, J. D., Easter, R. C., Gustafson Jr, W. I., Liu, X., Ghan, S. J.,  
19 and Singh, B.: Assessing the CAM5 physics suite in the WRF-Chem model:  
20 implementation, evaluation, and resolution sensitivity, *Geosci. Model Dev. Discuss.*, 6,  
21 6157-6218, 10.5194/gmdd-6-6157-2013, 2013.

22 Mashayekhi, R., and Sloan, J. J.: Effects of aerosols on precipitation in north-eastern  
23 North America, *Atmos. Chem. Phys.*, 14, 5111-5125, 10.5194/acp-14-5111-2014, 2014.

24 Matsui, H., Koike, M., Kondo, Y., Moteki, N., Fast, J. D., and Zaveri, R. A.:  
25 Development and validation of a black carbon mixing state resolved three-dimensional  
26 model: Aging processes and radiative impact, *J. Geophys. Res.*, 118, 2304-2326,  
27 10.1029/2012jd018446, 2013.

28 McKeen, S., Chung, S. H., Wilczak, J., Grell, G., Djalalova, I., Peckham, S., Gong, W.,  
29 Bouchet, V., Moffet, R., Tang, Y., Carmichael, G. R., Mathur, R., and Yu, S.: Evaluation  
30 of several PM<sub>2.5</sub> forecast models using data collected during the ICARTT/NEAQS 2004  
31 field study, *J. Geophys. Res.*, 112, D10S20, 10.1029/2006jd007608, 2007.

32 Morrison, H., Curry, J. A., and Khvorostyanov, V. I.: A New Double-Moment  
33 Microphysics Parameterization for Application in Cloud and Climate Models. Part I:  
34 Description, *J. Atmos. Sci.*, 62, 1665-1677, 10.1175/jas3446.1, 2005.

35 Morrison, H., Thompson, G., and Tatarskii, V.: Impact of Cloud Microphysics on the  
36 Development of Trailing Stratiform Precipitation in a Simulated Squall Line: Comparison

1 of One- and Two-Moment Schemes, *Mon. Wea. Rev.*, 137, 991-1007,  
2 10.1175/2008mwr2556.1, 2009.

3 Neale, R. B., Chen, C.-C., Gettelman, A., Lauritzen, P. H., Park, S., Williamson, D. L.,  
4 Conley, A. J., Garcia, R., Kinnison, D., Lamarque, J. F., Marsh, D., Mills, M., Smith, A.  
5 K., Tilmes, S., Vitt, F., Morrison, H., Cameron-Smith, P., Collins, W. D., Iacono, M. J.,  
6 Easter, R. C., Ghan, S. J., Liu, X., Rasch, P. J., and Taylor, M. A.: Description of the  
7 NCAR Community Atmosphere Model (CAM 5.0), NCAR, 2012.

8 Ntelekos, A. A., Smith, J. A., Donner, L., Fast, J. D., Gustafson, W. I., Chapman, E. G.,  
9 and Krajewski, W. F.: The effects of aerosols on intense convective precipitation in the  
10 northeastern United States, *Quarterly Journal of the Royal Meteorological Society*, 135,  
11 1367-1391, 10.1002/qj.476, 2009.

12 Pfister, G. G., Avise, J., Wiedinmyer, C., Edwards, D. P., Emmons, L. K., Diskin, G. D.,  
13 Podolske, J., and Wisthaler, A.: CO source contribution analysis for California during  
14 ARCTAS-CARB, *Atmos. Chem. Phys.*, 11, 7515-7532, 10.5194/acp-11-7515-2011,  
15 2011.

16 Qian, Y., Gong, D., Fan, J., Leung, L. R., Bennartz, R., Chen, D., and Wang, W.: Heavy  
17 pollution suppresses light rain in China: Observations and modeling, *J. Geophys. Res.*,  
18 114, n/a-n/a, 10.1029/2008jd011575, 2009.

19 Saide, P. E., Spak, S. N., Carmichael, G. R., Mena-Carrasco, M. A., Yang, Q., Howell,  
20 S., Leon, D. C., Snider, J. R., Bandy, A. R., Collett, J. L., Benedict, K. B., de Szoek, S.  
21 P., Hawkins, L. N., Allen, G., Crawford, I., Crosier, J., and Springston, S. R.: Evaluating  
22 WRF-Chem aerosol indirect effects in Southeast Pacific marine stratocumulus during  
23 VOCALS-REx, *Atmos. Chem. Phys.*, 12, 3045-3064, 10.5194/acp-12-3045-2012, 2012.

24 Shrivastava, M., Lane, T. E., Donahue, N. M., Pandis, S. N., and Robinson, A. L.: Effects  
25 of gas particle partitioning and aging of primary emissions on urban and regional organic  
26 aerosol concentrations, *J. Geophys. Res.*, 113, n/a-n/a, 10.1029/2007jd009735, 2008.

27 Shrivastava, M., Fast, J., Easter, R., Gustafson Jr, W. I., Zaveri, R. A., Jimenez, J. L.,  
28 Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of  
29 simple and complex representations of the volatility basis set approach, *Atmos. Chem.*  
30 *Phys.*, 11, 6639-6662, 10.5194/acp-11-6639-2011, 2011.

31 Shrivastava, M., Berg, L. K., Fast, J. D., Easter, R. C., Laskin, A., Chapman, E. G.,  
32 Gustafson, W. I., Liu, Y., and Berkowitz, C. M.: Modeling aerosols and their interactions  
33 with shallow cumuli during the 2007 CHAPS field study, *J. Geophys. Res.*, 1343-1360,  
34 10.1029/2012jd018218, 2013.

35 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G.,  
36 Huang, X.-Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research  
37 WRF Version 3, NCAR/NCAR/TN-475+STR, 2008.

1 Solomon, S., Qin, D., Manning, M., Alley, R. B., Bernsten, T., Bindoff, N. L., Chen, Z.,  
2 Chidthaisong, A., Gregory, J. M., Hegerl, G. C., Heimann, M., Hewitson, B., Hoskins, B.  
3 J., Joos, F., Jouzel, J., Kattsov, V., Lohmann, U., Matsuno, T., Molina, M., Nicholls, N.,  
4 Overpeck, J., Raga, G., Ramaswamy, V., Ren, J., Rusticucci, M., Somerville, R., Stocker,  
5 T. F., Whetton, P., Wood, R. A., and Wratt, D.: Technical Summary., in: *Climate Change*  
6 *2007: The Physical Science Basis. Contribution of Working Group I to the Fourth*  
7 *Assessment Report of the Intergovernmental Panel on Climate Change*, edited by:  
8 Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M.,  
9 and Miller, H. L., Cambridge University Press, Cambridge, United Kingdom and New  
10 York, NY, USA, 2007.

11 Stevens, B., and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a  
12 buffered system, *Nature*, 461, 607-613, 2009.

13 Stull, R. B.: A fair-weather cumulus cloud classification scheme for mixed-layer studies,  
14 *J. Climate Appl. Meteor.*, 24, 49-56, 1985.

15 Taylor, G. R.: Sulfate Production and Deposition in Midlatitude Continental Cumulus  
16 Clouds. Part I: Cloud Model Formulation and Base Run Analysis, *J. Atmos. Sci.*, 46,  
17 1971-1990, 10.1175/1520-0469(1989)046<1971:spadim>2.0.co;2, 1989.

18 Tremblay, A., and Leighton, H.: A Three-Dimensional Cloud Chemistry Model, *J.*  
19 *Climate Appl. Meteor.*, 25, 652-671, 10.1175/1520-  
20 0450(1986)025<0652:atdccm>2.0.co;2, 1986.

21 Tsimpidi, A. P., Karydis, V. A., Zavala, M., Lei, W., Molina, L., Ulbrich, I. M., Jimenez,  
22 J. L., and Pandis, S. N.: Evaluation of the volatility basis-set approach for the simulation  
23 of organic aerosol formation in the Mexico City metropolitan area, *Atmos. Chem. Phys.*,  
24 10, 525-546, 10.5194/acp-10-525-2010, 2010.

25 Vilà-Guerau de Arellano, J., Kim, S. W., Barth, M. C., and Patton, E. G.: Transport and  
26 chemical transformations influenced by shallow cumulus over land, *Atmos. Chem. Phys.*,  
27 5, 3219-3231, 10.5194/acp-5-3219-2005, 2005.

28 Wang, C., and Chang, J. S.: A three-dimensional numerical model of cloud dynamics,  
29 microphysics, and chemistry: 1. Concepts and formulation, *J. Geophys. Res.*, 98, 14827-  
30 14844, 10.1029/92jd01393, 1993.

31 Wang, H., Rasch, P. J., and Feingold, G.: Manipulating marine stratocumulus cloud  
32 amount and albedo: a process-modelling study of aerosol-cloud-precipitation interactions  
33 in response to injection of cloud condensation nuclei, *Atmos. Chem. Phys.*, 11, 4237-  
34 4249, 10.5194/acp-11-4237-2011, 2011.

35 Wang, H., Easter, R. C., Rasch, P. J., Wang, M., Liu, X., Ghan, S. J., Qian, Y., Yoon, J.  
36 H., Ma, P. L., and Vinoj, V.: Sensitivity of remote aerosol distributions to representation  
37 of cloud-aerosol interactions in a global climate model, *Geosci. Model Dev.*, 6, 765-782,  
38 10.5194/gmd-6-765-2013, 2013.

- 1 Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando,  
2 J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global  
3 model to estimate the emissions from open burning, *Geosci. Model Dev.*, 4, 625-641,  
4 10.5194/gmd-4-625-2011, 2011.
- 5 Wyngaard, J. C.: Toward Numerical Modeling in the “Terra Incognita”, *J. Atmos. Sci.*,  
6 61, 1816-1826, 10.1175/1520-0469(2004)061<1816:tnmitt>2.0.co;2, 2004.
- 7 Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol  
8 Interactions and Chemistry (MOSAIC), *J. Geophys. Res.*, 113, D13204,  
9 10.1029/2007jd008782, 2008.
- 10 Zhang, G. J., and McFarlane, N. A.: Sensitivity of climate simulations to the  
11 parameterization of cumulus convection in the Canadian climate centre general  
12 circulation model, *Atmosphere-Ocean*, 33, 407-446, 10.1080/07055900.1995.9649539,  
13 1995.
- 14 Zhang, Y., Sartelet, K., Wu, S. Y., and Seigneur, C.: Application of WRF/Chem-  
15 MADRID and WRF/Polyphemus in Europe – Part 1: Model description, evaluation of  
16 meteorological predictions, and aerosol–meteorology interactions, *Atmos. Chem. Phys.*,  
17 13, 6807-6843, 10.5194/acp-13-6807-2013, 2013.
- 18 Zhao, C., Liu, X., Ruby Leung, L., and Hagos, S.: Radiative impact of mineral dust on  
19 monsoon precipitation variability over West Africa, *Atmos. Chem. Phys.*, 11, 1879-1893,  
20 10.5194/acp-11-1879-2011, 2011.
- 21 Zhao, C., Chen, S., Leung, L. R., Qian, Y., Kok, J. F., Zaveri, R. A., and Huang, J.:  
22 Uncertainty in modeling dust mass balance and radiative forcing from size  
23 parameterization, *Atmos. Chem. Phys.*, 13, 10733-10753, 10.5194/acp-13-10733-2013,  
24 2013.

25  
26  
27

1 Table 1. WRF-Chem configuration used in this study

Physical Process	Parameterization
Surface	Noah land-surface model (Chen et al., 1996)
Boundary layer	Mellor-Yamada-Janjić (Janjić, 1990, 2002)
Cloud microphysics	Morrison two moment (Morrison et al., 2005; Morrison et al., 2009)
Cumulus	Kain-Frisch (with CuP modifications) (Kain and Fritsch, 1990; Kain, 2004; Berg et al., 2013)
Radiation (shortwave and longwave)	CAM 3 (Collins et al., 2004)
Gas-phase chemistry	SAPRC-99 (Carter, 2010)
Aqueous Chemistry	Fahey and Pandis (Fahey and Pandis, 2001; Chapman et al., 2009).
Aerosol chemistry	MOSAIC for inorganic aerosols (Zaveri et al., 2008); Simplified Volatility Basis Set (VBS) for organic aerosol (Shrivastava et al., 2011). Eight size bins, dry diameters 0.039-10 $\mu\text{m}$

2

3

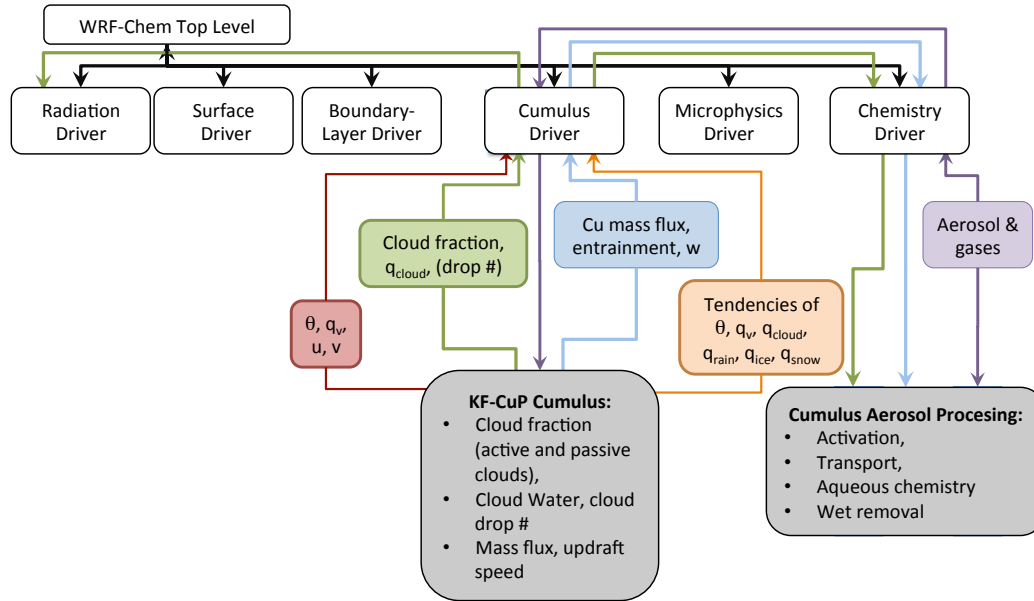
- 1 Table 2. Definitions of simulations completed as part of the study. The parameterized  
2 cumulus dynamics are applied in all simulations.

Simulation	Aerosol Processing by Shallow and Deep Cu	Status (on/off)
DeepShallow	Aerosol Processing Shallow Cu	<b>On</b>
	Aerosol Processing Deep Cu	<b>On</b>
ShallowOnly	Aerosol Processing Shallow Cu	<b>On</b>
	Aerosol Processing Deep Cu	Off
Control	Aerosol Processing Shallow Cu	Off
	Aerosol Processing Deep Cu	Off

3

4

# 1 Figures



2

3 Figure 1. Summary of modifications to the standard implementation of WRF-Chem.

4 Colored boxes indicate information passed between subroutines related to the

5 thermodynamics (red), cloud microphysical and macrophysical properties (green), cloud

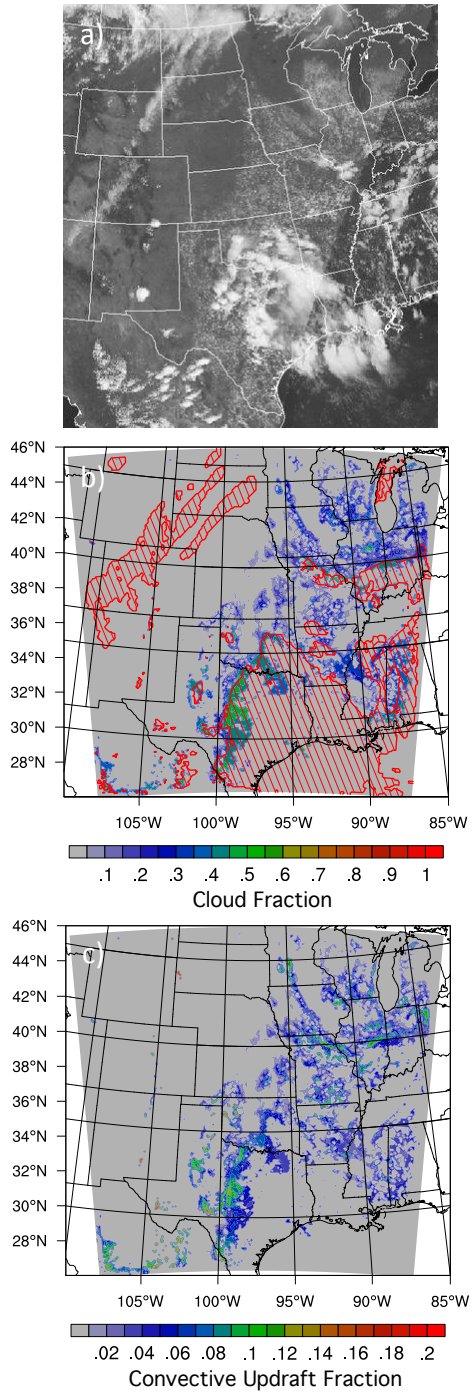
6 dynamics (blue), thermodynamic tendencies (orange) and aerosol and trace gases

7 (purple), while gray boxes indicate the new or modified parameterizations applied in

8 WRF-Chem. Arrows indicate information flow within the model. Note that the droplet

9 number generated in the KF-CuP parameterization is not currently used in the Radiation

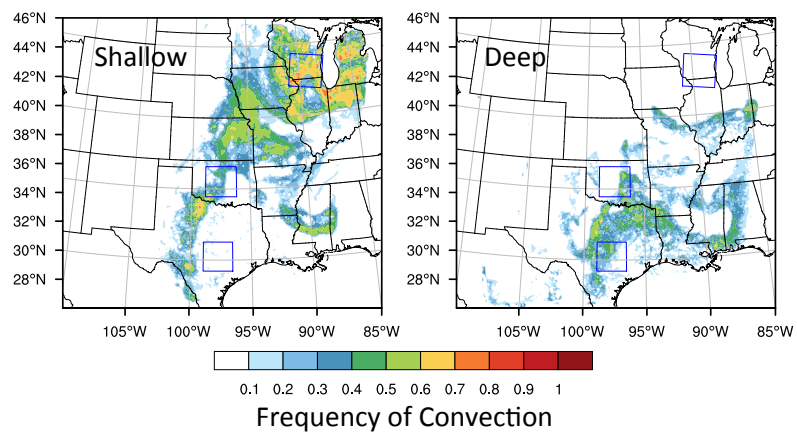
10 Driver.



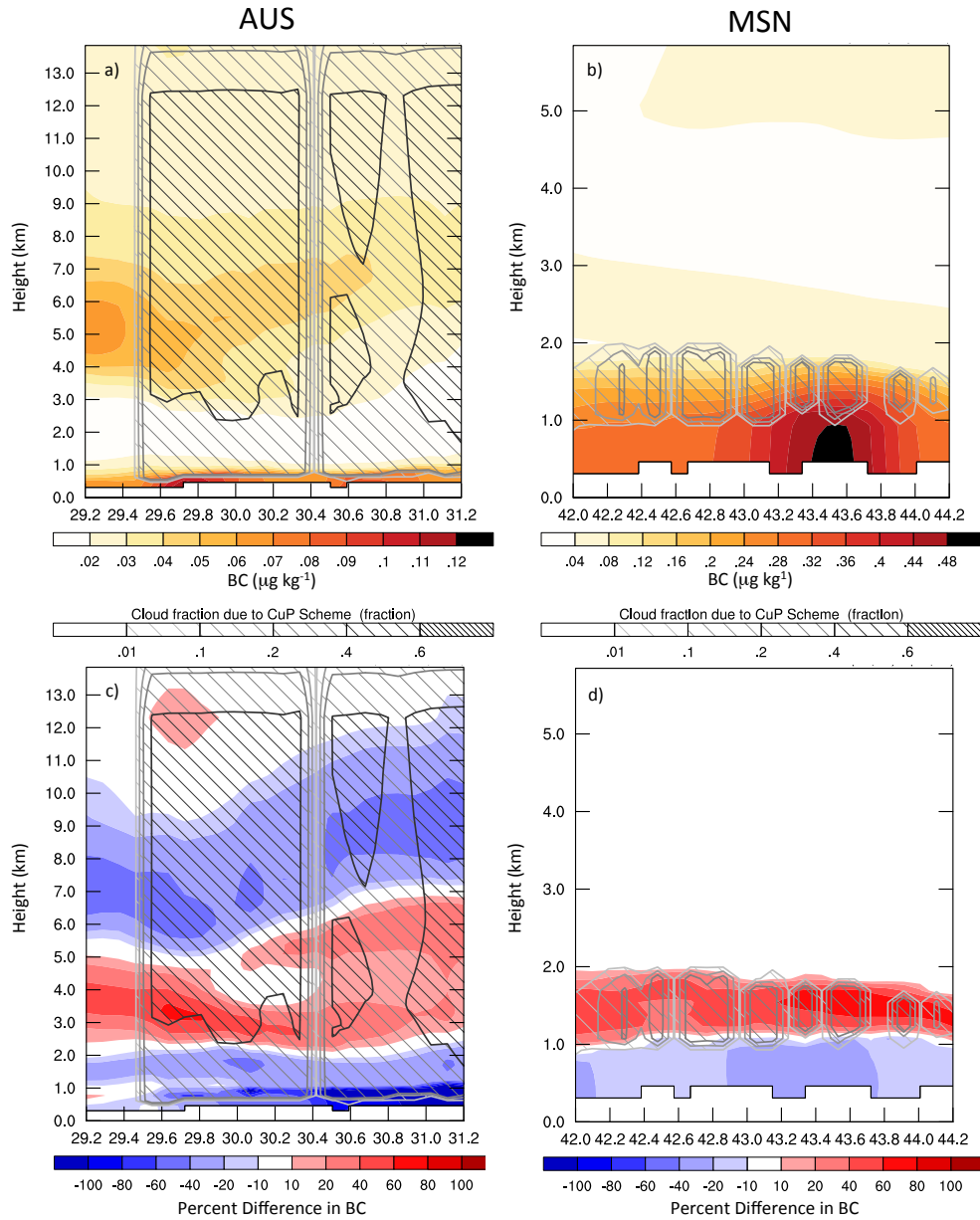
1

2 Figure 2. GOES visible satellite image valid at 20:15 UTC, 25 June 2007 (a), and  
 3 simulated cloud fraction associated with the KF-CuP parameterization (colors), areas  
 4 with grid resolved clouds (hashed; b), and fraction of model grid box with convective  
 5 updrafts (c). Note different color scales used in plots of cloud fraction (b) and convective  
 6 updraft fraction (c).

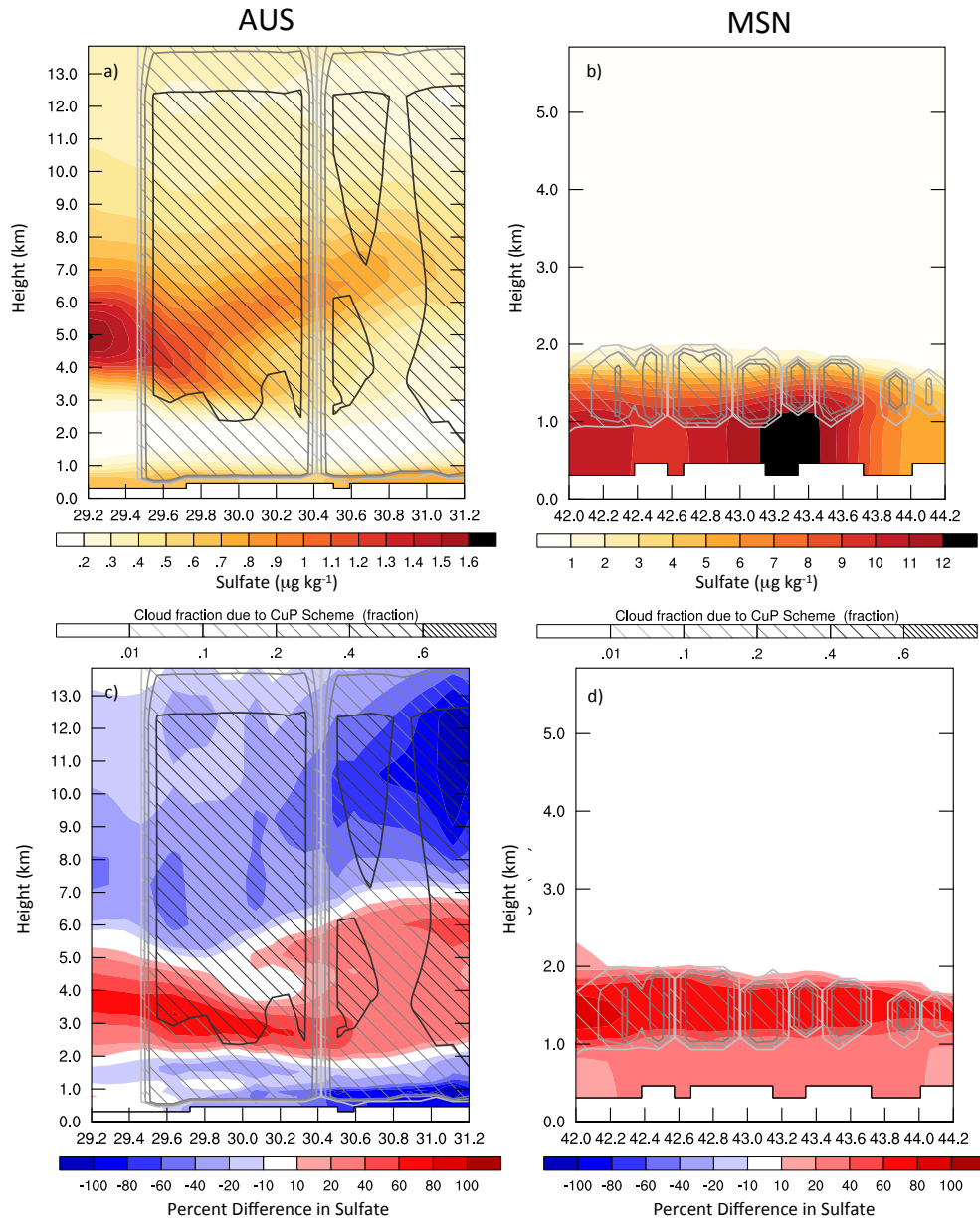




1  
2 Figure 3. Frequency of occurrence of deep convection (right) and shallow convection  
3 (left) for the time period 12:00-20:00 UTC on 25 June, 2007. Boxes indicate sub regions,  
4 240 km on a side, selected for analysis.

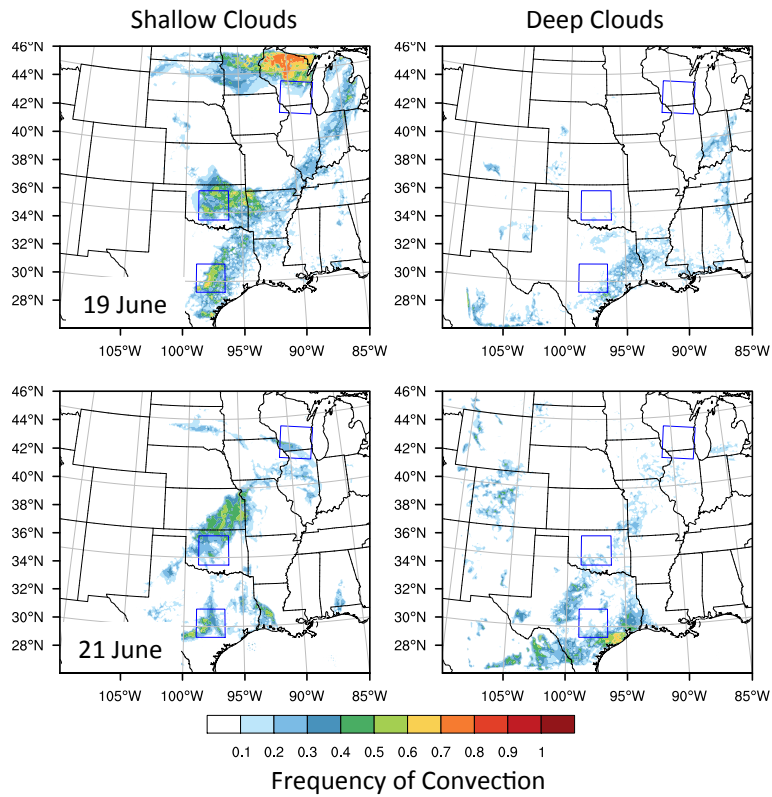


1  
2 Figure 4. Vertical north-south cross sections of BC **mixing ratio summed over** size bins 1  
3 through 4 [**color shading in a) and b);  $\mu\text{g kg}^{-1}$** ], including both interstitial and activated  
4 aerosol in the cloudy grid cells, and difference in BC **mixng ratio** between DeepShallow  
5 and control simulations [**color shading in c and d); percentage**] for conditions dominated  
6 by deep convective clouds [AUS; **a) and c)**] and shallow convective clouds [MSN; **b) and**  
7 **d)**] boxes at 20 UTC on 25 June, 2007. Hatching indicates cloud fraction associated with  
8 sub-grid convective clouds. The horizontal axes **are** labeled in degrees of latitude and **the**  
9 **vertical axes are** height above mean sea level.



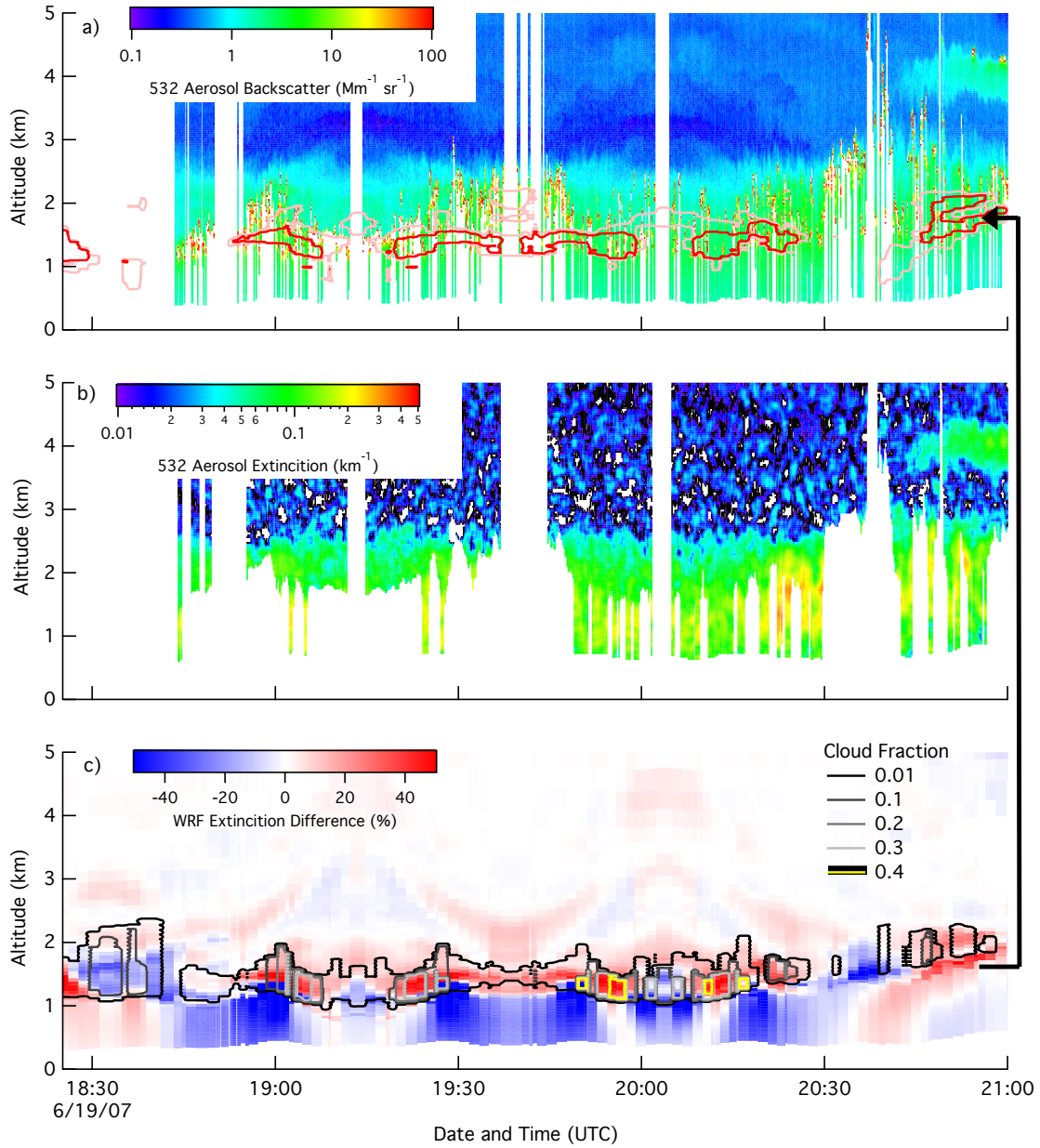
1

2 Figure 5. Same as Figure 4, but for sulfate.

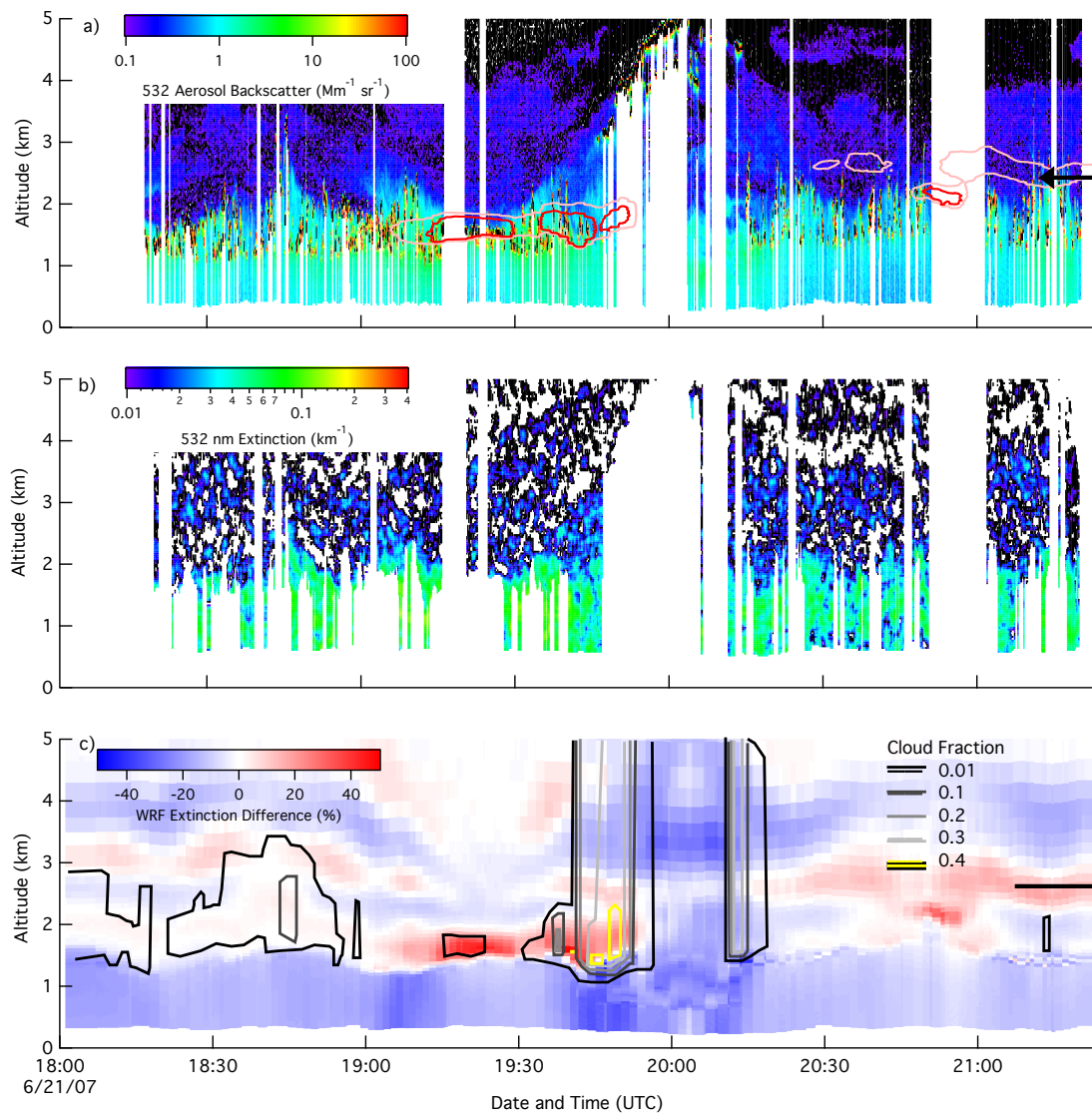


1

2 Figure 6. Frequency of occurrence of deep convection (right) and shallow convection  
 3 (left) for the time period 12:00-20:00 UTC on 19 (top) and 21 June (bottom), 2007.



1  
2 Figure 7. Cross sections of observed aerosol backscatter (a), aerosol extinction (b) at  
3 wavelength of 532 nm, and difference in aerosol extinction of DeepShallow and Control  
4 simulations (c) on 19 June 2007. Contours in (a) mark contours of +10 and +20%  
5 difference in the WRF-Chem simulations, as indicated by the large arrow, and contours  
6 in (c) indicate simulated cloud fraction as indicated by the legend.



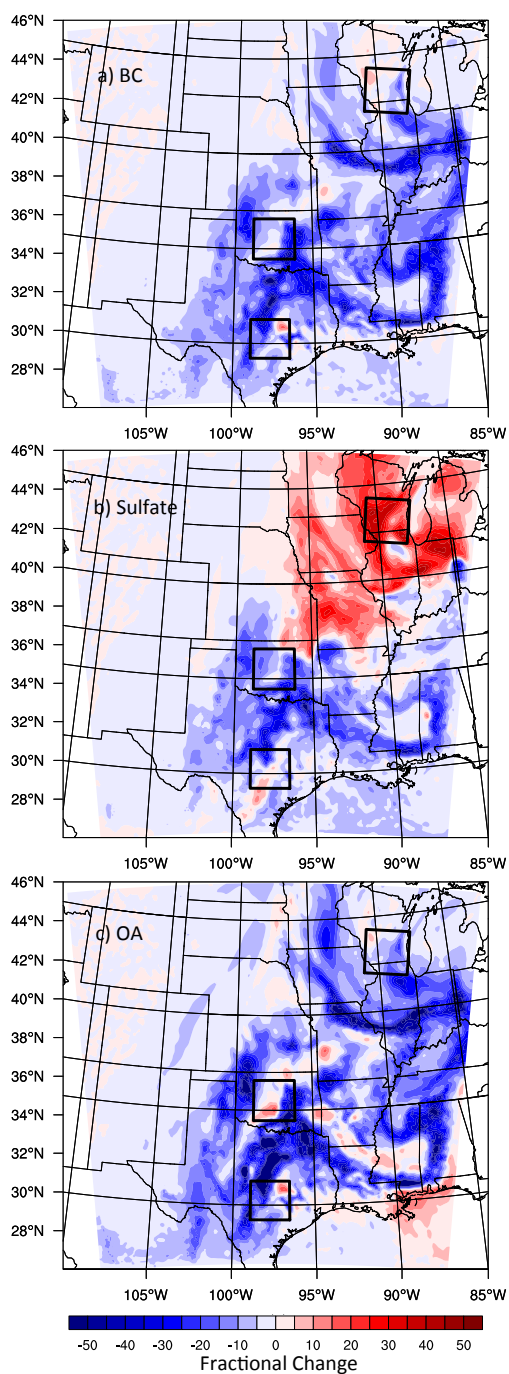
1

2 Figure 8. Same as Figure 7 but for 21 June 2007.

3



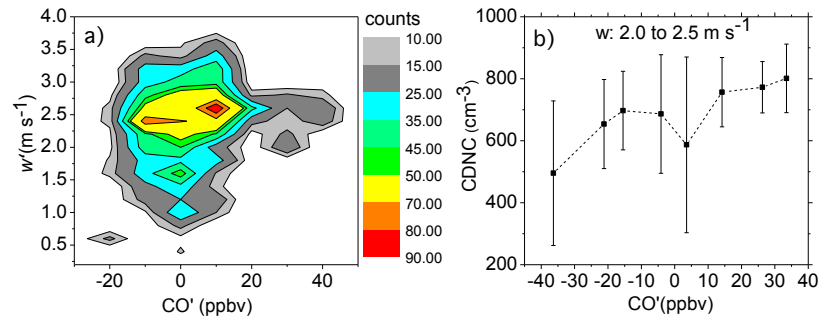
1



2

3 Figure 9. Fractional differences in column integrated aerosol mass loading between  
 4 DeepShallow and control simulations for size bins 1 through 4, including both interstitial  
 5 and activated aerosol in the cloudy grid cells, for BC (a), sulfate (b) and OA (c), valid at  
 6 20:00 UTC on 25 June, 2007. Yellow boxes indicate boxes used in the analysis.

1  
2



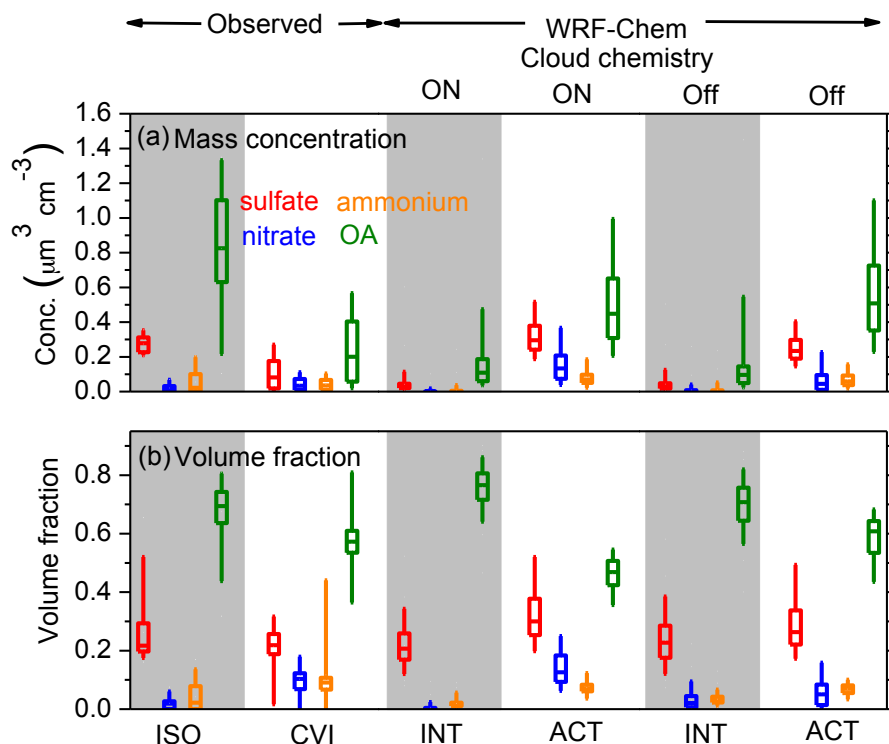
3

4 Figure 10. PDF of simulated cloud updraft speed and CO loading in cloudy updrafts (a),  
5 change in CDNC with perturbation values of CO ( $\text{CO}'$ ) for perturbation values of  $w$  ( $w'$ )  
6 between 2.0 and 2.5  $\text{ms}^{-1}$  (b). Error bars in (b) indicate the standard deviation.

7

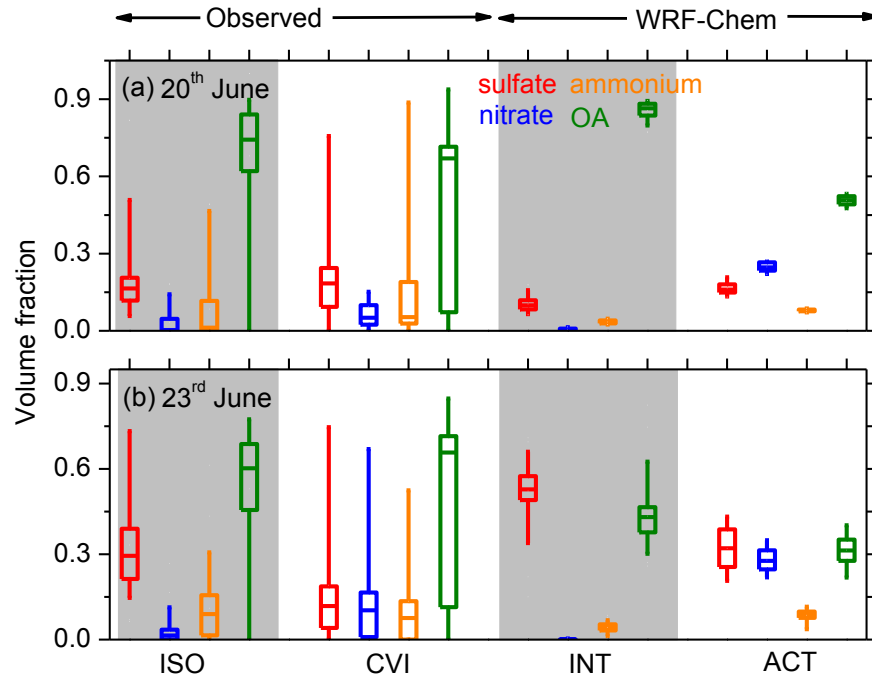


1



2

3 Figure 11. Aerosol mass concentration (a) and volume fraction (b) for observed  
 4 interstitial [sampled via an isokinetic inlet (ISO; grey areas)] and activated [sampled via a  
 5 counter-flow virtual impactor inlet (CVI; white areas)] aerosol; and simulated interstitial  
 6 (INT; grey areas) and activated (ACT; white areas) aerosol at 20:00 UTC on 25 July,  
 7 2007. Colors indicate sulfate (red), ammonium (orange), nitrate (blue), and organic  
 8 aerosol (green) in size bins 1 through 4. Box-and-whisker plots indicate 90<sup>th</sup>, 75<sup>th</sup>, 50<sup>th</sup>,  
 9 25<sup>th</sup>, and 10<sup>th</sup> percentiles.



1

2 Figure 12. Similar to Figure 11, but for only aerosol volume fraction on 20 (a) and 23 (b)  
 3 June 2007. WRF-Chem results are only for cases in which the aqueous chemistry is  
 4 turned on.

5