Anonymous Referee #1

I find this manuscript much improved over the previous version. I appreciate that the authors responded actively to most of the critical comments that I made in my earlier review.

I am still worrying about the single case simulation to derive the conclusion. I would like to see the simulation results for other fog cases besides Tule fog case, because we do not know how the modified SOWC model simulates other fog cases under different meteorological condition and how the modified radiation scheme affects the fog formation. The authors just speculate the expected results as following ("Thus, for other fog cases the results might be different because the model can perform differently, better or worse, but we expect that the conclusion will be similar, if fog is successfully simulated"). However, I will leave the decision to the editor.

Reply: The authors appreciate the reviewer's suggestions. We would very much like to extend our study to fog cases under different weather conditions over other regions to have a more robust conclusion on how aerosol mixing states affect fog formation and the radiation budget. However, to run a case at a different region requires a significant amount of effort. Aerosols in California have been carefully investigated using the SOWC model (Joe et al., 2014; Zhang et al., 2014). Thus the model gives more creditability to study a Tule fog event in California. While the WRF model has been well tested under different weather conditions over different regions in the world, we still need reliable emission data and reasonable air pollution simulations over a new region before we study a fog event there. Since the project ended, we do not have resources to extend the air pollution work over another region during a fog event. Without a careful study verifying that the emissions data support robust air pollution predictions over a new region using the SOWC model, we are very reluctant to draw conclusions about a fog case there. We are working to secure additional funding for these future studies, but they are beyond the scope of the current work. We have added a comment in the revised manuscript summary to reflect this. Lines 665-667.

"Note that the current results are based on a Tule fog case study in the Central Valley of California. Additional fog case studies under different weather conditions in other regions of the world are required to draw conclusions at those locations."

Anonymous Referee #2

This paper investigates the effect of aerosol mixing state on fog formation by implementing warm-cloud processes in a source-oriented WRF/Chem model. With several model experiments, the authors make great efforts to show that the source-oriented representation of particles is a more realistic approach than the internal mixture approach for the study of direct, indirect, and semi-indirect effects of anthropogenic aerosols. The topic of the paper is definitely of crucial importance for studying aerosol–cloud–radiation interactions and for predicting the effects of climate change on the hydrological cycle and energy budget. However, the advantages of the source-oriented approach are not fully applied in this study although the results of aging processes are compared with those of internal mixing state assumption. The specific mechanism of aerosol influence on fog formation is not well investigated as it should be. Moreover, the model used in this study underestimated ammonium nitrate aerosols significantly, and the impacts on predicted hygroscopic properties of aerosols and thus CCN should have been investigated. The paper needs to be more concise and better written. Below are my detailed comments (for version2 of the manuscript), which need to be addressed before the paper can be considered for publication.

Reply: The authors appreciate the reviewer's comments. Follows are point-by-point response to the reviewer's comments. The bolded texts are added in the revised manuscript according to reviewer's comments.

Major comments:

1. Sect. 2.1, Sect. 2.2, Fig. 1 and Table 1: The source-oriented representation of aerosol particles (AQC) as well as fog droplets (CLDAQC) is declared to be the core part of this study. However, whereas the AQC number concentrations are presented in Table 6, other sourced-oriented variables are not shown and actually none of the AQC and CLDAQC variables is given in the figures of this paper. In fact, the results of "source-oriented" simulations are not explicitly investigated in the paper. The "source-oriented" model appears not to be a fully suitable expression although it is frequently used in the paper; otherwise, more analyses of the source-oriented simulation results should be performed.

The authors appreciate the reviewer's suggestion using "aging-process-included" instead of "sourced-oriented" to represent aerosol activation under near-realistic mixing condition in minor comment 4. We modified some phrases in the revised version to clarify the meaning. However, the term "source-oriented" is used in Zhang et al. (2014) to indicate the aerosol mixing state and chemistry presentation in the modified WRF/Chem model (i.e., the source-oriented WRF/Chem, SOWC, model). Since the current manuscript follows on to the studies from Zhang et al. (2014), we have retained the term "sourced-oriented" in the revised manuscript.

2. The authors did several model experiments, as defined in Table 3, to investigate how aerosol mixing state influences fog formation. (a) However, they did not outlet clearly what are the specific mechanisms of aerosol influence on fog formation based on the above model results. (b) For the statement in Line 276-282, what are the other

microphysical processes the authors mean? It would be very helpful if all the steps of aerosol-cloud-radiation interactions could be described here for clouds as well as for fogs. Please formulate the mechanism carefully for the model results and discussion.

(a) In the previous version of the manuscript, the discussion about how aerosol mixing state influences fog formation is scattered around Section 4.3. In this revised version, we provide a brief summary and discussion about how aerosol mixing state influences fog formation in the end of Section 4.3.
"In summary, compared to S_ARon_CRmod, I_ARon_CRmod has a higher CCN / cloud droplet number concentration because internally mixed aerosols can instantaneously contain hydroscopic material (e.g. sulfate) through artificial mixing, which decreases the critical supersaturation requirement for a particle to activate into a CCN, leading to higher cloud number concentration and optical thickness. Thicker fog in I_ARon_CRmod reduces the amount of shortwave radiation reaching the surface, resulting in a lower surface temperature. A lower surface temperature can have a positive feedback on cloud lifetime (i.e., a longer cloud lifetime), which further reduces shortwave radiation reaching the surface when compared to S_ARon_CRmod. Hence, the aerosols that include aging processes can delay CCN activation and produce fewer cloud droplets and less fog, which in turn will modify the energy budget near the surface." in Lines 618-628.

(b) The brief concept of aerosol-cloud-radiation interactions has been introduced in the Introduction Section, Lines 70-76. In cloud microphysics, once aerosol particles activate as CCNs and become cloud droplets, they grow in size by diffusion initially. Later when droplets grow larger, autoconversion and collision/coalescence will occur to form rain droplets. If the ice phase (i.e., ice, snow, and graupel) exists, more microphysical processes will be involved, such as the Bergeron effect, riming, aggregation, melting, etc. Besides activation, the evaporation from rain droplets or melting from ice particles can contribute to the number of cloud droplets. We have modified the text to
"Fog is an excellent scientific case study that can isolate cloud activation and diffusive growth, the first step of aerosol-cloud-radiation interactions, from other microphysical processes that usually do not occur in fog, such as collision/coalescence, riming, melting, and aggregation. This paper presents the development of the CLDAQC treatment within the SOWC model to ensure that the model performs properly. A careful selection of a weather phenomenon is important to evaluate the model performance and the impact of the aerosol mixture states on cloud formation. The involvement of other hydrometeors, in addition to cloud droplets, can not only change the cloud properties and cloud lifetime, but also modify the energy budget. These extremely nonlinear processes can significantly complicate the evaluation of the model performance and the first step of aerosol-cloud-radiation interaction. Thus we choose fog as our very first weather system study for the evaluation of aerosol-cloud-radiation interactions using the improved SOWC model." in Lines 276-287.

3. In the S_ARon_CRmod scheme, the cloud droplets are allowed to range in size from dry aerosol particle radius to 30 microns when calculating the cloud optical thickness (Line 479-480). This setting is likely to result in errors when calculating the cloud optical thickness, because cloud droplet radius is usually greater than 1

microns and thus unacted particles are involved in the calculation. Please state clearly the reasons for this setting.

The sentence that we wrote was misleading and we have clarified the description in the revised version. We do not include unactivated aerosols in the calculation of cloud optical thickness (COT). In the modified radiation scheme (e.g., S_ARon_CRmod), cloud droplet size varies for each bin and source types. In the original radiation scheme, the effective radius (r_e) is diagnosed assuming a constant radius which is capped within a range (4 μm ≤ r_e ≤ 20 μm). In our modified code, we use simulated cloud radii to directly calculate COT and dry aerosol radii are only used for the lower bound of cloud droplet sizes to avoid numerical error (i.e. division by zero). Our calculation of the COT includes cloud droplets of all sizes (even very small ones), but the COT calculation does not include unactivated aerosols. We have modified the sentence to "**In the modified cloud-radiation scheme (CRmod), the size range of cloud droplets in Eq. (3) can vary between activated CCNs to 30 μm.**" in Lines 362-364.

4. As described in Line 483 and later on in the paper, the simulation results of optical thickness between S_ARon_CRmod and S_ARon_CRorig are very similar. The authors think "because the meteorological conditions of the fog event are calm and stable, the cloud microphysics processes are fairly slow and simple (no rain produced in this case)". However, from the simulations of mean values of cloud liquid water (NOT mixing ratio as stated in the paper) and cloud droplet number (Table 5), one can see that the cloud water mixing ratios are similar, but the cloud droplet number concentrations are quite different, which means that the mean cloud droplet sizes should be different between the two experiments. Cloud droplet number concentration, cloud droplet size and cloud droplet index of refraction are the factors that can affect the calculation results of cloud optical thickness. So the authors need to synthesize the above-mentioned factors, and give the specific reasons for the simulation results.

This is a good question. We have described the differences of the cloud optical thickness calculation between S_ARon_CRorig and S_CRon_CRmod in response to reviewer's comments in the previous revision. Since more than one reviewer has raised the same question, we have added the following in Lines 496-508 of the revised version.

"**In the original radiation scheme, the cloud optical thickness (COT) is a function of cloud water path (CWP) and an effective radius (4 μm ≤ r_e ≤ 20 μm), which is derived from the water mass and the total droplet number assuming a uniform size of cloud droplets:**

$$\tau_{orig}(\lambda) = CWP \times (-6.59 \times 10^{-3} + \frac{1.65}{r_e}). \quad (6)$$

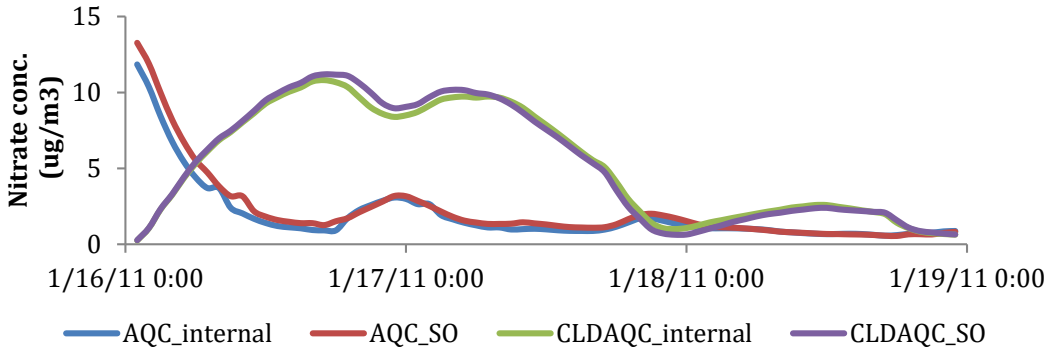
In the modified radiation scheme in S_ARon_CRmod, the COT is calculated based on simulated cloud droplet size, number, and chemical composition of each bin and source (Eq. 3). In addition, the formula of COT, single scattering albedo and asymmetry factor in the modified radiation scheme are all modified. With a similar Qc, although Qn in S_ARon_CRorig is higher than that in S_ARon_CRmod, the COT is slightly higher in S_ARon_CRmod due to different formulas used in the calculation of cloud-radiation interaction. The small

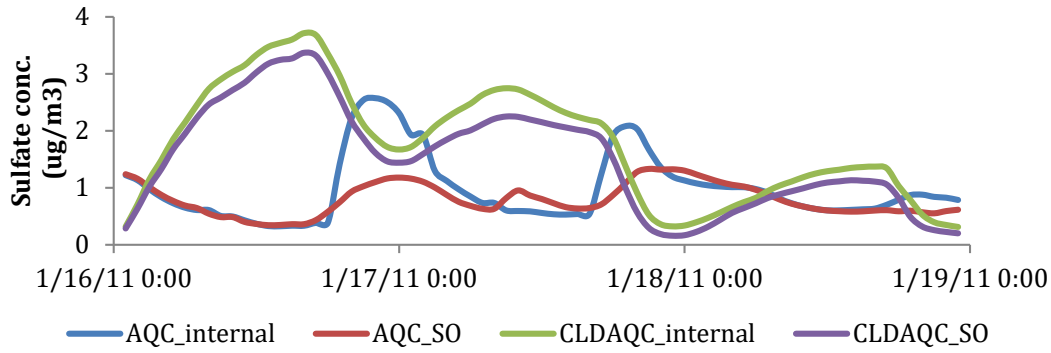
difference of COT between these two experiments in fact indicates that the parameterization of COT in the original radiation scheme provides a reasonable result compared to the explicit COT calculation.“

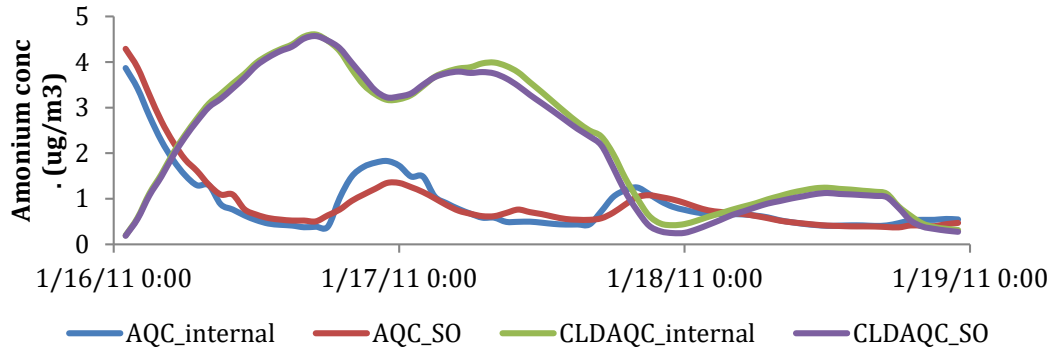
We thank the reviewer pointing out the incorrect use of the term “cloud water mixing ratio”. We correct the term to “cloud liquid water” in the revised version.

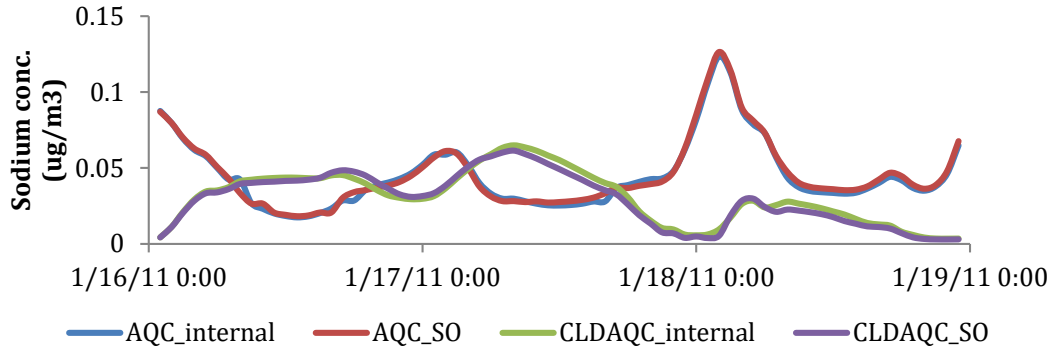
5. As described in Sect. 4.1 of the paper (Line 399-412), both simulated nitrate and ammonium concentrations are underestimated by 70%. This implies that ammonium nitrate, which has a great potential of absorbing water molecules, is significantly underestimated. What effects of such poor behavior of the model on the size distributions and hydroscopic properties of aerosols, CCN and fog formation? It would not be advised if the reader is merely referred to a paper of Zhang et al. (2014) without providing the cause of discrepancy and the effects on fog simulation.

The nitrate under-prediction is not unusual in reactive chemical transport models (e.g., WRF/Chem and CMAQ; see for example peak nitrate under-predictions in Fig 6 of Kelly et al. (2014)). There are numerous current research projects underway to identify and correct the cause of this problem. We had hoped that feedbacks between meteorology and atmospheric chemistry were part of the answer and that is the hypothesis that was tested in the current study. Unfortunately, the feedbacks do not seem to increase predicted nitrate formation, and so we must rely on results from other projects to address this point. However, based on the time series of chemical concentration in aerosols (AQC) and cloud (CLDAQC) in S_ARon_CRmod and I_ARon_CRmod (figures below), the model well simulated nucleation process for four species. Although we do not know whether the Nitrate and Ammonium concentration were underestimated before the fog formed, our model did correctly simulate aerosol activation processes and the concentration of those chemical species can be tracked in both aerosols (AQC) and cloud (CLDAQC). Nitrate and Ammonium were under-predicted after fog dissipation on 18 January because most Ammonium and Nitrate are released back to the gas phase. This potentially explains why Nitrate and Ammonium are significantly underestimated in the condensed phase (Fig. 6 in the manuscript). Unfortunately, we only have observations on the last day of simulation (18th) when fog almost dissipated. With this limited observations, it is difficult to effectively evaluate the cause of the underestimation of Nitrate and Ammonium.









Minor comments:

1) Line 90-93: In fact, the aging timescale was investigated previously based on aircraft measurements, e.g., in the work of Ma et al. (2010). Ma et al. (2010) showed that the coating of dust particles by high pollution acids (due to high emission rates of their precursors) is very efficient with a timescale of a few hours, providing the predominant source of CCN, and such coating is more efficient with increasing OH (which is a measure of the oxidation capacity of the atmosphere). Therefore, instead of stating that little information is available, such kind of work should be cited in the paper.

The authors thank the reviewer for providing the useful information and suggestion. We have modified the text to **"From the IPAC-NC (Influence of Pollution on Aerosols and Cloud Microphysics in North China) field campaign data, Ma et al. (2010) showed that dust particles can become the predominant source of CCNs in a few hours after being coated by high pollution acids. This aging process has**

been parameterized in numerical models (Lesins et al., 2002) but how the aging timescale should respond to changes in temperature, humidity, oxidant concentrations and/or emissions rates is not described in most models." in Lines 104-110.

2) Line 107-113 and Line 163-164: Does the model include new particle formation processes? It looks that the new nucleation mode (typically for particle sizes less than 25 nm) is not taken into account in both the schemes. It is stated that the particle size of first bin from emissions is 5 nm, which appears too small compared to the size of the first bin (39 nm) the model scheme has considered.

The authors thank the reviewer to point out the typo in the manuscript. The particle size of the first bin in our model is 55 nm, not 5 nm. Both MADE-SORGAM and MOSAIC aerosol schemes in the WRF/Chem include new particle formation processes and so does SOWC but these processes were not active in the current simulations. The San Joaquin Valley is one of the most heavily polluted air basins in the United States with relatively large concentrations of primary particles but at the same time relatively low emissions of SO_2 due to emissions control programs that limit the sulfur content of fuel. Under these conditions, nucleation of new particles is a minor process compared to emissions and atmospheric processing of primary particles. The current manuscript focuses on these mechanisms, and defers the discussion of nucleation to a future study in a location where this process is important for CCN formation.

3) Line 178-193: There are several cloud schemes in WRF-Chem, such as Morrison scheme. Why do the authors select Purdue Lin scheme, and do you have verify the simulation capability of the scheme?

The Purdue Lin scheme was developed at Purdue University and was published by the second author of the manuscript and her advisor (Chen and Sun, 2002). Thus we are very familiar with the scheme and the code. The scheme has been used in many studies (Cruz and Narisma, 2016; Anber et al., 2016; and many others). Although the original Purdue Lin scheme is a one-moment scheme, Chapman et al. (2009) added a prognostic treatment of cloud droplet number to the scheme to make it a two-moment scheme on cloud water within WRF/Chem. This is the first scheme in the WRF model that considers the aerosol-cloud interaction (CCN). From the reasons mentioned above, we therefore chose this scheme for the study.

4) Line 280: It might be more suitable to use a phrase like "aging-process-included" to replace the "source-oriented" here, as the basic chemical and physical processes for fog formation are the same no matter whether the sources are tagged or not.

We agree that using "aging-process-included" instead of "sourced-oriented" in this sentence is more appropriate to represent aerosol activation under near-realistic mixing condition. We have modified this instance and similar instances of the phrase throughout the revised manuscript to clarify the meaning. Again, the term "source-oriented" is used in Zhang et al. (2014) to indicate our new modified treatment of aerosol chemistry mixing state in the model (the source-oriented WRF/Chem, SOWC). Since this manuscript follows on to the studies by Zhang et al., we prefer to retain the term "source-oriented" in appropriate locations throughout the manuscript.

5) Line 282: What instruments were used to measure surface relative humidity? Could the fog formation take place below RH=100% in the Central Valley (see e.g. Kulmala et al., 1997; Makkonen et al., 2012)? By using what parameters and their threshold values is a fog event identified to occur in this study?

A hygrometer is an instrument used to measure relative humidity in most ground-based weather stations, which belong to National Weather Service. Based on reviewer's suggestion, we have added "**For example, the IPAC-NC field campaign in China observed clouds formed in a polluted environment with RH below 100% due to high hydroscopic pollutants (Ma et al., 2010).**" in Lines 293-294.

RH measurements were just one of the parameters used to select the current case for study with the SOWC model. We also used geostationary satellite images as another observation of fog conditions. Due to some high hydroscopic pollutants (e.g., Nitrate) formed in the Central Valley, we believe it may be possible that Tule fog can form when RH is below 100%, though we do not have data to verify.

6) Line 416-417: What causes are for such an underestimation of LWP? Could this be liked to an underestimation of ammonium nitrate?

The cause for the underestimation of LWP in the middle portion of the Central Valley is not clear. The third author has different projects working on this research area. However, as we descripted above, underestimated Nitrate and Ammonium is probably a consequence of fog formation (i.e., released back to the gas phase after evaporation) in this study.

7) Line 466-467: How or by what processes does the aerosol direct effect lead to increases of the cloud water mass and cloud droplet number?

Aerosol particles absorb and scatter shortwave radiation, leading to a smaller shortwave flux reaching the surface. In our study, Table 5 shows that the average NSF within the Central Valley from S_ARoff_CRmod is higher than from S_ARon_CRmod by 3.7 W m^{-2}. This means that the aerosol direct radiative forcing reduces an average of the shortwave energy flux on the ground by 3.7 W m^{-2} and thus produces a lower skin temperature in this case study. A lower skin temperature (0.1 K) provides a favorable condition for cloud/fog formation near the surface. Compared to S_ARoff_CRmod, the mean values of cloud liquid water and cloud droplet number are increased by 3.3% and 4.5%, respectively, in S_ARon_CRmod (Table 5).

8) There have been some studies about the effect of aerosol mixing state on CCN and warm cloud formation before this work. The author should refer to the literatures during their analysis and discussion (e.g., Cubison et al., 2008; Lance et al.,2013; Anttila et al., 2010;).

We have added the following in the Introduction Section of the revised manuscript.

"**Some studies showed the importance of the aerosol mixing state on CCN activation using field campaign data and numerical modeling. Cubison et al. (2008) focused on the relationship between the CCN number concentration and**

the physical and chemical properties of aerosols in the urban area in California. Based on the results from a cloud parcel model and observed CCN number, they found that a realistic treatment of the mixing state of the urban aerosol distribution is critical to the CCN activation prediction. Anttila (2010) also used an adiabatic cloud parcel model to investigate the importance of the particle mixing state and hygroscopicity to CCN activation. They commented that the differences between externally and internally mixed aerosols in urban and rural environments could reach up to 35%. Both modeling studies investigated the effect of aerosol mixing state on CCN formation based on an ideal cloud parcel model. While observations from Ma et al. (2010) and Lance et al. (2013) showed consistent results with these modeled CCN studies, a more sophisticated 3-dimational numerical model is needed to further study this issue." in Lines 88-101.

Technical issues:

Line 430-431: Smaller cloud droplets are not shown in the figure.

Observed LWP and modeled LWP are comparable with one-day shift (Fig. 7). Based on Eq. (3), to obtain a higher cloud optical thickness one expects to have a higher number concentration with smaller cloud droplets in order to maintain a comparable LWP. We have rewritten the sentence to clarify this point.

"While simulated LWP is comparable to MODIS retrievals with one-day shift (Fig. 7), to obtain a higher COT than observed (Fig. 8b versus 8c) we expect that the model produces more small cloud droplets with a higher CCN concentration, especially over highly polluted areas. " in Lines 449-451.

Formulas (2)-(5), Tables 4 and 5: Same character Q is used for different variables though having different subscripts.

Q_e, Q_s, Q2, Q_c, and Q_n are the dimensionless extinction efficiency, the dimensionless scattering efficiency, 2-m water vapor mixing ratio, cloud liquid water, and cloud droplet number, respectively. Q always has a subscript and should therefore be clear to the reader.

Line 463, 574, 589: Should "water mixing ratio" be a dimensionless variable or it should be called water content?

We corrected "cloud water mixing ratio" to "cloud liquid water" in the revised version.

Line 544-545: Can one learn hydrophobic nature of particles from Table 6?

We have modified the sentences to "**Most particles tracked in the AQC size bin 1 can activate in the internally mixed experiment; however, in the source-oriented experiment only particles emitted in AQC size bin 1 from wood smoke and "other sources" are sufficiently hygroscopic to activate. The remaining sources are dominated by hydrophobic compounds (such as elemental carbon) that do not activate under the study conditions (Table 6).**" in Lines 564-568.

Line 555-557: How one can see the changes after late 17 January from Table 4 that shows only mean values over 16-18 January?

Figure 9 shows time series of mean bias variation of 2-m temperature, 2-m vapor mixing ratio, and NSF from different experiments. Thus we revised the sentence to state "**After late 17 January, the bias differences between two experiments are more apparent in the daytime than in the nighttime (Fig. 9 and Table 4).**" in Lines 558-559.

Line 617: better to use important "results", instead of "findings".

Modified.

Table 1: not fully useful.

Table 1 was added based on another reviewer's comment to provide the aerosol chemistry information for readers. We prefer to retain the table based on that original reviewer suggestion.

Table 6: What does the Ratio mean? Information on particle size ranges of each bin can be provided.

The ratio is the aerosol concentration for each bin/source divided by the total. This has been clarified in the caption of Table 6. The size range for each AQC bin (S_ARon _CRmod) is now added in the supplementary.

"Table 6. The ratio of AQC number concentration for each bin/source to the total number concentration. The numbers are averaged within the first five model layers during 16 to 18 January 2011."

(Units in μm)

bin1	0.058	~	0.069
bin2	0.122	~	0.139
bin3	0.239	~	0.260
bin4	0.480	~	0.507
bin5	0.874	~	1.070
bin6	0.914	~	1.886
bin7	2.633	~	4.541
bin8	5.695	~	10.844

Line 1091 (Fig. 7 legend): should be (b), (d) and (f).

Corrected.

Line 1123 (Fig. 10): At what supersaturation?

It is real ratio of activated CCN number to existing aerosol number. So the supersaturation varies at each grid point.

Line 1154 (Fig. 13): better be plotted in colour.

Changed.

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Implementation of Warm-Cloud Processes in a Source-Oriented WRF/Chem Model to Study the Effect of Aerosol Mixing State on Fog Formation in the Central Valley of California

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Abstract

The source-oriented Weather Research and Forecasting chemistry model (SOWC) was modified to include warm cloud processes and applied to investigate how aerosol mixing states influence fog formation and optical properties in the atmosphere. SOWC tracks a 6-dimensional chemical variable (X, Z, Y, Size Bins, Source Types, Species) through an explicit simulation of atmospheric chemistry and physics. A source-oriented cloud condensation nuclei module was implemented into the SOWC model to simulate warm clouds using the modified two-moment Purdue Lin microphysics scheme. The Goddard shortwave and longwave radiation schemes were modified to interact with source-oriented aerosols and cloud droplets so that aerosol direct and indirect effects could be studied.

The enhanced SOWC model was applied to study a fog event that occurred on 17 January 2011, in the Central Valley of California. Tule fog occurred because an atmospheric river effectively advected high moisture into the Central Valley and nighttime drainage flow brought cold air from mountains into the valley. The SOWC model produced reasonable liquid water path, spatial distribution and duration of fog events. The inclusion of aerosol-radiation interaction only slightly modified simulation results since cloud optical thickness dominated the radiation budget in fog events. The source-oriented mixture representation of particles reduced cloud droplet number relative to the internal mixture approach that artificially coats hydrophobic particles with hygroscopic components. The fraction of aerosols activating into CCN at a supersaturation of 0.5% in the Central Valley decreased from 94% in the internal mixture model to 80% in the source-oriented model. This increased surface energy flux by 3-5 W m^{-2} and surface temperature by as much as 0.25 K in the daytime.

1. Introduction

Atmospheric aerosols are complex mixtures of particles emitted from many different anthropogenic and natural sources suspended in the atmosphere. In contrast to greenhouse gases, aerosols have large spatial and temporal variability in the troposphere because of their short lifetimes (about one week) before coagulation, dry deposition, or wet scavenging processes remove them from the atmosphere (Ramanathan et al., 2001). Aerosol particles can influence human health (McMichael et al., 2006), ecological health (over land and ocean) (Griffin et al., 2001), visible range through the atmosphere , cloud / precipitation formation (Chen et al., 2008), and the net radiation budget of the earth (IPCC, 2007). Some chemical components of aerosol particles are important to direct radiative forcing of the climate due to their optical properties (Tegen et al., 1996). Particulate sulfate scatters incoming solar radiation, leading to an estimated direct forcing of -0.95 W m^{-2} (Adams et al., 2001). Particulate black carbon strongly absorbs incoming shortwave radiation, which warms the mid-level of the atmosphere but cools the earth's surface (Yang et al., 2009; Koch and Del Genio, 2010). Particulate black carbon also leads to reduce relative humidity and cloud liquid water content (semi-direct effect) in the mid-level atmosphere (Ackerman et al., 2000; Koch and Del Genio, 2010). In addition to these direct effects, Twomey (1974) proposed that aerosols indirectly affect the earth's energy budget due to their ability to serve as cloud condensation nuclei (CCN), which are of great importance in cloud development, especially for warm clouds in the mid-to-high latitudes. Large numbers of CCN produce clouds with a greater number of smaller size cloud droplets (Chen et al., 2008). These smaller cloud droplets raise cloud albedo (the first indirect effect) and also suppress the formation of precipitation and prolong cloud lifetime (the second indirect effect) (Albrecht, 1989). The direct, semi-direct, and indirect effects of aerosol particles modify the energy budgets in the atmosphere and on the surface, with corresponding changes in atmospheric stability. The

2007 IPCC report (IPCC, 2007) concluded that the net forcing of all aerosols could be either positive or negative in the range from -0.7 W m^{-2} to +0.1 W m^{-2}. The majority of this uncertainty is associated with the semi-direct and indirect effects due to the complexity of aerosol-cloud interactions.

The magnitude of the aerosol semi-direct and indirect effects depends on the number concentration, size, and composition of the atmospheric aerosol particles that act as CCN or ice nuclei (IN) (Lohmann and Feichter, 2005; Chen et al., 2008). Particles with hygroscopic components such as water-soluble ions (Na^+, Cl^-, SO_4^{2-}, NO_3^- etc.) readily act as CCN (Chen and Lamb, 1994). Particles that contain hydrophobic components such as freshly emitted organic carbon or elemental carbon must become coated with hygroscopic material before they will easily serve as CCN (Dusek et al., 2006). This aging process is often parameterized in models (Lesins et al., 2002) but little information is available to describe how the aging timescale should respond to changes in temperature, humidity, oxidant concentrations and/or emissions rates. Mineral dust particles (Motoi, 1951; Georgii and Kleinjung, 1967) commonly have a favorable arrangement of surface structure that allows them to serve as IN. Secondary coatings that condense on mineral dust particles may reduce their ability to serve as IN (Sullivan et al., 2010) but increase their ability to serve as CCN (Li and Shao, 2009). All of these effects. Some studies showed the importance of the aerosol mixing state on CCN activation using field campaign data and numerical modeling. Cubison et al. (2008) focused on the relationship between the CCN number concentration and the physical and chemical properties of aerosols in the urban area in California. Based on the results from a cloud parcel model and observed CCN number, they found that a realistic treatment of the mixing state of the urban aerosol distribution is critical to the CCN activation prediction. Anttila (2010) also used an adiabatic cloud parcel model to investigate the importance of the particle mixing state and hygroscopicity to CCN activation. They commented that the differences

between externally and internally mixed aerosols in urban and rural environments could reach up to 35%. Both modeling studies investigated the effect of aerosol mixing state on CCN formation based on an ideal cloud parcel model. While observations from Ma et al. (2010) and Lance et al. (2013) showed consistent results with these modeled CCN studies, a more sophisticated 3-dimational numerical model is needed to further study this issue. Mineral dust particles (Motoi, 1951; Georgii and Kleinjung, 1967) commonly have a favorable arrangement of surface structure that allows them to serve as IN. Secondary coatings that condense on mineral dust particles may reduce their ability to serve as IN (Sullivan et al., 2010) but increase their ability to serve as CCN (Li and Shao, 2009). From the IPAC-NC (Influence of Pollution on Aerosols and Cloud Microphysics in North China) field campaign data, Ma et al. (2010) showed that dust particles can become the predominant source of CCNs in a few hours after being coated by high pollution acids. This aging process has been parameterized in numerical models (Lesins et al., 2002) but how the aging timescale should respond to changes in temperature, humidity, oxidant concentrations and/or emissions rates is not described in most models. All of these mentioned above point to the importance of the particle mixing state when predicting CCN / IN concentrations.

The standard Weather Research and Forecasting (WRF) model, including the chemistry component (WRF/Chem), permits the simulation of the combined direct, indirect and semi-direct effects of aerosols (Chapman et al., 2009; Fast et al., 2006; Grell et al., 2005). WRF/Chem Version 3.1.1 has sophisticated packages to represent chemistry processes (i.e. gas-phase reaction, gas-to-particle conversion, coagulation, etc.) and aerosol size and composition (Zaveri et al., 2008; Ackermann et al., 1998; Binkowski and Shankar, 1995; Schell et al., 2001). The Modal Aerosol Dynamics Model for Europe with Secondary Organic Aerosol Model (MADE-SORGAM) and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) are commonly used aerosol schemes in the

~~WRF/Chem model. Both schemes have inorganic, organic, and secondary organic aerosols and contain aerosol formation processes including nucleation, condensation, and coagulation. The main difference between MADE SORGAM and MOSAIC is the representation of aerosol size distributions. MADE SORGAM uses 3 log normal modes (Aitken, accumulation and coarse) while MOSAIC uses 4 (or 8) aerosol size sections (bins) from 39 nm to 10 μm, respectively. The details of MADE SORGAM are described in Binkowski and Shankar (1995), Ackermann et al. (1998), Schell et al. (2001), and Grell et al. (2005) and the details of MOSAIC are given in Zaveri et al. (2008).~~

~~As mentioned above, the size, composition, and mixing state of aerosols strongly affect their ability to activate into cloud droplets~~ . The Modal Aerosol Dynamics Model for Europe with Secondary Organic Aerosol Model (MADE-SORGAM) and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) are commonly used aerosol schemes in the WRF/Chem model. Both schemes have inorganic, organic, and secondary organic aerosols and contain aerosol formation processes including nucleation, condensation, and coagulation. The main difference between MADE-SORGAM and MOSAIC is the representation of aerosol size distributions. MADE-SORGAM uses 3 log-normal modes (Aitken, accumulation and coarse) while MOSAIC uses 4 (or 8) aerosol size sections (bins) from 39 nm to 10 μm, respectively. The details of MADE-SORGAM are described in Binkowski and Shankar (1995), Ackermann et al. (1998), Schell et al. (2001), and Grell et al. (2005) and the details of MOSAIC are given in Zaveri et al. (2008).

As mentioned above, the size, composition, and mixing state of aerosols strongly affect their ability to activate into cloud droplets (Lance et al., 2013; Zaveri et al., 2010). However, most WRF/Chem chemistry packages make a global internal mixing assumption in which all particles within a log-normal mode (MADE-SORGAM) / size bin (MOSAIC scheme) in the

same grid cell are instantaneously combined such that they have the same chemical composition. In reality, airborne particles are emitted with unique chemical composition and only become internally mixed over a period of hours to days depending on atmospheric conditions. The instantaneous internal mixing assumption alters the optical and chemical properties of particles in WRF/Chem simulations (Zhang et al., 2014) and therefore has the potential to influence aerosol-cloud interaction (i.e. CCN activation).

The primary goal of this research is to quantify the effect of assumptions about particle mixing state on predicted cloud droplet formation within the WRF/Chem model. Warm cloud processes in the Purdue Lin scheme (Chen and Sun, 2002) were modified in the Source-Oriented WRF/Chem (SOWC) model to investigate the impact of aerosol mixing state on the characteristics of a fog event in the Central Valley of California. The SOWC model explicitly predicts particle mixing state in the presence of emissions, transport, coagulation, chemical transformation, and deposition. The integration of warm-cloud processes with the source-oriented treatment of particles in the current study provides a more realistic approach to understand how mixing state influences direct, indirect, and semi-indirect effects of anthropogenic aerosols.

This paper is organized as follows: the model description and development of warm cloud processes are introduced in section 2; observational data and numerical experiment design are presented in section 3; results are discussed in section 4; and the summary and discussion are provided in section 5.

2. Model Description and Development

2.1 SOWC

WRF is a compressible, non-hydrostatic regional meteorology model, which uses the Arakawa C grid and terrain-following hydrostatic pressure coordinates. The governing

equations of the model are written in flux form and can be solved using a range of solution schemes. In the present study, the Runge-Kutta third-order time scheme was employed and fifth- and third-order advection schemes were chosen for the horizontal and vertical directions, respectively (Skamarock et al., 2008). WRF/Chem simulates trace gas and particle chemical concentrations concurrently with the meteorological fields using the same grid structure, the same advection scheme, and the same physics schemes for sub-grid scale transport (Grell et al., 2005). The SOWC model was developed based on WRF/Chem V3.1.1 with significant modifications throughout the code to enable the use of 6D variables. The standard WRF/Chem model tracks 3-dimensionial chemistries in a 4-dimensional variable (X, Z, Y, Species). The SOWC model tracks a 6-dimensional chemical variable "AQC" (X, Z, Y, Size Bins, Source Types, Species). Particles emitted from different sources have different sizes and chemical compositions, leading to a source-oriented mixture of particles that age in the atmosphere through coagulation and gas-particle conversion (e.g., condensation and evaporation) processes. Airborne particles in the SOWC model influence meteorological conditions through radiative feedbacks and microphysical processes. The model simultaneously tracks particle mass, number concentration, and radius. The number concentration and radius of different particle size bins from each source type are included as the last two elements in the species dimension. Simulations in this study use 38 chemical species (Table 1) from 5 emissions sources (wood smokes, gasoline, diesel, meat cooking, and other aerosol types) and 8 size bins. The initial particle sizes from emissions are 0.~~005~~055, 0.1105, 0.221, 0.4415, 0.8835, 1.767, 3.535, and 7.0693 microns. Note that the SOWC model uses moving size bins whose sizes change in response to gas-particle conversion during model simulations. The model conserves aerosol mass concentration throughout the simulation of atmospheric processes including emissions, transport, deposition, coagulation, and condensation/evaporation. The gas-phase species emitted from

different sources in each grid cell are not tracked separately in the SOWC model at the present time. In the current study, the initial and boundary conditions of aerosol particles are based on observations from the California Regional Particulate Air Quality Study (CRPAQS) (Ying et al., 2008). The distribution of particle emissions for different bins for every source are calculated using emissions inventories provided by the California Air Resources Board (CARB) along with measured chemical speciation profiles (Ying et al., 2008). Further details of the SOWC model structure and source-oriented chemistry processes are described by Zhang et al. (2014) and Joe et al. (2014).

2.2 Cloud microphysics scheme

The original Purdue Lin microphysics scheme was designed as a one-moment water mass conserved microphysics scheme with five hydrometeors: cloud water, rain, cloud ice, snow, and graupel (Lin et al., 1983; Chen and Sun, 2002). Chapman et al. (2009) added a prognostic treatment of cloud droplet number (Ghan et al., 1997) to the Purdue Lin scheme to make a two-moment treatment of cloud water within WRF/Chem. In our study, a source-oriented CCN module was added to the SOWC model to track size-resolved information about activated CCN from various aerosol sources. A new source-oriented 6D cloud variable, "CLDAQC" (X, Z, Y, Size Bins, Source Types, Species) was added to SOWC to describe source-oriented clouds. Droplet radius and number concentration are once again stored as the last two elements in the species dimension of the CLDAQC variable. In the Purdue Lin scheme, all microphysics processes are parameterized with water mass, except autoconversion. ~~Chapman et al. (2009) added the autoconversion parameterization from Liu et al. (2005) into the Purdue Lin microphysics, which depends on cloud droplet number. Chapman et al. (2009)~~Chapman et al. (2009) added the autoconversion parameterization from Liu et al. (2005) into the Purdue Lin microphysics, which depends on cloud droplet number. Chapman et al. (2009) also specified changes to cloud droplet number proportional to the

microphysics process rate of cloud water mass. For example, when 10% cloud water becomes rain water after autoconversion, 10% cloud droplets will be moved at the same time.

The continuity equation of the mass-coupled mixing ratio of CLDAQC can be written as follows:

$$\frac{\partial CLDAQC}{\partial t} = \nabla \cdot \vec{V} CLDAQC + \nabla \cdot K \nabla CLDAQC + P_{AACT} + S_{micro}, \quad (1)$$

where $\vec{V}$ is the 3D wind vector and K is the eddy diffusion coefficient. The first two terms on the right hand side of Eq. (1) are the flux divergence of CLDAQC (transport) and sub-grid eddy mixing, respectively. Figure 1 shows the schematic diagram of the sinks and sources of CLDAQC in the cloud microphysics processes (P_{AACT} and S_{micro}). Aerosol activation (P_{AACT}) is the main source of CLDAQC. The calculation of aerosol activation is based on a maximum supersaturation determined from a Gaussian spectrum of updraft velocities and aerosol chemistry composition for each size bin (Abdul-Razzak and Ghan, 2002). This parameterization of aerosol activation was implemented in WRF/Chem model (Chapman et al., 2009) and is used in this study. Aerosol activation was calculated each time step. Once the environment reached the critical supersaturation, AQC activated as CCN. Water vapor condenses at a diffusion limited rate to cloud droplets (water molecules transferred from vapor to cloud in Purdue Lin scheme) and particle mass/number is transferred from the interstitial aerosol variable (AQC) to the cloud-borne aerosol variable (CLDAQC). The Purdue Lin microphysics scheme uses a saturation adjustment approach (i.e., it adjusts water vapor to the saturation mixing ratio), so CCN activation is calculated before saturation adjustment. After saturation adjustment, the condensation rate due to vapor diffusion is proportional to particle size (Rogers and Yau, 1989). Results from CCN activation tests at relevant supersaturation are discussed in Section 4.3.

Sinks and sources of CLDAQC (S_{micro}) are based on interactions between a cloud droplet and the other hydrometeors (e.g., ice, rain, snow, and graupel) that can remove water from or add water to CLDAQC. The sinks of cloud water, as well as CLDAQC, include autoconversion from cloud to rain (P_{RAUT}) and the accretion of cloud water by rain (P_{RACW}), snow (P_{SACW}), and graupel (P_{GACW}). The exchange between cloud water and cloud ice can also occur through homogenous freezing of cloud water to ice (P_{IHOM}) and melting of cloud ice to cloud water (P_{IMLT}). Finally, the condensation (associated with P_{ACCT}) and evaporation of cloud water (P_{CEVP}) are implicitly taken into account in the Purdue Lin microphysics scheme. When cloud droplets fully evaporate (sink of CLDAQC), the residual cores are released back into the corresponding source type and size bin of the aerosol (AQC) variable.

2.3 Radiation schemes

The NASA Goddard shortwave and longwave radiation schemes (Chou and Suarez, 1999b, 2001b) are used in conjunction with the source-oriented cloud droplet algorithms in the enhanced SOWC model. Absorption of radiation by water vapor, ozone, oxygen, carbon dioxide, cloud droplets and aerosol particles is considered. Interactions among the absorption and scattering by clouds and aerosols (Mie scattering), molecules (Rayleigh scattering) and the surface are fully accounted for (Skamarock et al., 2008). Three main optical parameters are calculated for each model layer to describe the influence of aerosols on the radiation: aerosol optical thickness (τ), single scattering albedo (ω), and asymmetry factor (g). In the present study, the numerical code described by Ying and Kleeman (2003) was implemented to calculate the optical properties of source-oriented particles. The original numerical code of Mie scattering developed by Bohren and Huffman (1983) was used to calculate the particle extinction efficiency, scattering efficiency and asymmetry factor. The partial molar refractive index approach described in Stelson (1990) was used to estimate the mean refractive index for multi-component aerosols. .

For any wavelength of shortwave or longwave radiation (λ), the aerosol optical thickness (τ_a) of a model layer with depth h (m) containing a number concentration $n_a(r)$ (# m^{-3} μm^{-1}) of droplets with radius r (μm) is given by

$$\tau_a(\lambda) = \pi h \int_0^\infty Q_e(\lambda, r) r^2 n_a(r)\, dr, \quad (2)$$

where, Q_e is the dimensionless extinction efficiency. The equivalent definition of aerosol optical thickness for discrete size bins j with a mean radius r_j (μm) can be written as

$$\tau_a(\lambda) = \pi h \sum_i^n \sum_j^m Q_{ei,j}(\lambda, r) {r_{i,j}}^2 N_{i,j}, \quad (3)$$

where subscript i refers to emission source, subscript j refers to size, n is the number of particle source types and m is the number of particle sizes. N (# m^{-3}) is the number of particles. The mean asymmetry factor (g_a) and single scattering albedo (ω_a) are calculated using the method described in (Yang, 2000):

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$$g_a(\lambda) = \frac{\sum_i^n \sum_j^m Q_{si,j}(\lambda, r) g_{i,j}(\lambda, r) N_{i,j} \pi r_{i,j}^2}{\sum_i^n \sum_j^m Q_{si,j}(\lambda, r) N_{i,j} \pi r_{i,j}^2}, \quad (4)$$

$$\omega_a(\lambda) = \frac{\sum_i^n \sum_j^m Q_{si,j}(\lambda, r) N_{i,j} \pi r_{i,j}^2}{\sum_i^n \sum_j^m Q_{ei,j}(\lambda, r) N_{i,j} \pi r_{i,j}^2}, \quad (5)$$

where Q_s is the dimensionless scattering efficiency. All of the optical parameters are functions of the wavelength (λ) of incident radiation.

In the original Goddard radiation schemes, cloud droplets are assigned to a mono-disperse size distribution (mean effective radius) which depends on the water mass and number concentration. The source-oriented cloud (CLDAQC) contains size distribution and chemistry information which is more realistic than the mono-disperse assumption. Equations

3-5 are applied to all size bins of not only the AQC but also the CLDAQC variables to calculate optical properties and radiative forcing.

3. Numerical experiment designs

3.1 Fog event

A numerical simulation of fog was carried out with the SOWC model ~~as a convenient method~~ to test the effects of particle mixing state on warm clouds processes. Fog is an excellent scientific case study that can isolate cloud activation and diffusive growth, the first step of aerosol-cloud-radiation interactions, from other microphysical processes that usually do not occur in fog, such as collision/coalescence, riming, melting, and aggregation. This paper presents the development of the CLDAQC treatment within the SOWC model to ensure that the model performs properly. A careful selection of a weather phenomenon is important to evaluate the model performance and the impact of the aerosol mixture states on cloud formation. The involvement of other hydrometeors, in addition to cloud droplets, can not only change the cloud properties and cloud lifetime, but also modify the energy budget. These extremely nonlinear processes can significantly complicate the evaluation of the model performance and the first step of aerosol-cloud-radiation interaction. Thus we choose fog as our very first weather system study for the evaluation of aerosol-cloud-radiation interactions using the improved SOWC model. The influence of particle size and composition on fog formation and droplet growth has been studied in previous field experiments ~~(Frank et al., 1998;Moore et al., 2004;Ming and Russell, 2004;Cubison et al., 2008;Niu et al., 2012)~~(Frank et al., 1998; Moore et al., 2004; Ming and Russell, 2004; Cubison et al., 2008; Niu et al., 2012) and modeling studies ~~(Bott and Carmichael, 1993;Kleeman et al., 1997)~~(Bott and Carmichael, 1993; Kleeman et al., 1997). ~~The results indicate that particle chemical composition and mixing state strongly influence fog droplet activation, mirroring the processes of interest for cloud droplets.~~

. The results indicate that particle chemical composition and mixing state strongly influence fog droplet activation, mirroring the processes of interest for cloud droplets. For example, the IPAC-NC field campaign in China observed clouds formed in a polluted environment with RH below 100% due to high hydroscopic pollutants (Ma et al., 2010).

Tule fogs (radiation fog) frequently form in the Central Valley of California during the winter season (Hayes et al., 1992). Winter in the Central Valley is associated with the maximum concentration of airborne particulate matter (PM) (Chow et al., 1993) which is composed of aerosol particles that can act as CCN. We chose ~~this challenging weather system for the first study of this kind since~~ Tule fog as our case study since it is important in safety, hydrology and agriculture in California~~. Fog is also an excellent scientific case study that can isolate cloud activation~~ and ~~diffusive growth, the first step of aerosol-cloud-radiation interactions, from other microphysical processes which usually do not occur~~aerosols in ~~fog.~~California have been carefully investigated using the SOWC model (Joe et al., 2014; Zhang et al., 2014)~~.~~ In the present study, a thick fog event that occurred on 16 and 17 January 2011 (Fig. 2) was chosen to investigate the impact of ~~source-oriented~~aging-process-included aerosol-cloud-radiation interactions on fog formation.~~-~~ Fog started forming over the northern Central Valley on 13 January with observed surface relative humidity reaching 95-100% and extended to the southern Central Valley on 14 January. The fog became thicker on 16 January and reached the maximum on 17 January (Fig. 2). This is evident by retrieved cloud optical thickness from MODIS (discussed later). The fog started dissipating from the northern Central Valley on 18 January and fully dissipated on 19 January (Fig. 2c).

In addition to calm wind and radiative cooling, high moisture is an important ingredient to a Tule fog event in the Central Valley, California. Figure 3 shows the time series of column integrated water vapor, sea level pressure, and 850-hPa wind vectors from ECMWF Interim reanalysis data. On 11 January, the column water vapor (CWV) was very low, less

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than 10 mm, over the Central Valley (Fig. 3a). Moisture was advected into the Central Valley (Fig. 3b) by a winter cyclone moving close to the northwestern coast of the United Sates on 12 January. A weak southwest-northeast-oriented atmospheric river with a width of 1000 km and a maximum CWV of ~26-28 mm approached the western coast and brought moisture into the Central Valley. At 0000 UTC 13 January (Fig 3c), moisture content began increasing in the northern Central Valley. At night, drainage flow from the surrounding mountains brought cold air into the Central Valley, mixed with the low-level moist air, and initiated fog formation over the northern Central Valley. On 14 January (Fig. 3d), the CWV over the southern Central Valley reached 22-24 mm and fog formed over the southern Central Valley.

On 15 and 16 January, a more intense, west-southwest to east-northeast oriented atmospheric river advected moisture into northern California (Figs. 3e and f). The moisture in the Central Valley reached a maximum on 17 January (Fig. 3g), at the time when the fog reached its maximum thickness during the study period (Fig. 2; also see the cloud optical thickness discussion later). On 18 January (Fig. 3h), while high moisture and fog still presented over the southern Central Valley, the moisture decreased and the fog disappeared over the northern Central Valley. Fog fully dissipated in the Central Valley on 19 January.

According to the satellite images and surface temperature variation, the coverage and thickness of fog followed a diurnal pattern with thinning in the daytime and thickening at night. As mentioned earlier, the aerosol mixture state can impact fog formation and properties of cloud droplets.

3.2 Observational data

Multiple types of measurement data were used to evaluate the SOWC model performance. Moderate Resolution Imaging Spectroradiometer (MODIS) level 2 cloud products from the Terra and Aqua satellites provide 5-km resolution cloud optical thickness

(COT) and liquid water path (LWP). The LWP retrieval from MODIS has been used to study low cloud and fog (Bendix et al., 2005). High-resolution MODIS data can describe fog spatial distribution and intensity but are only available once every 24 hours (daytime only) from each satellite. The SOWC model predictions for temperature and moisture at the surface are also evaluated against *in situ* time-series meteorological data from 24 surface weather stations along with net ground shortwave fluxes at 42 sites from California Irrigation Management Information System (CIMIS). Measured concentrations of airborne particles were obtained from the California Ambient Air Quality Data (CAAQD) provided by the Planning & Technical Support Division (PTSD) of the California Air Resources Board (CARB). The station details of CAAQD are provided in Table 2. The locations of all measurement sites are provided in Fig. 4.

3.3 Numerical experiment design

The primary objective of this study is to examine how the source-oriented (S_) (i.e., aging-process-included) and internal (I_) mixture representations of aerosol particles differ in their feedbacks to meteorology in a fog event. Internally mixed simulations (I_) artificially blend emissions from all sources into a single particle size distribution thereby concealing all advanced treatments of particle mixing and aging. Four experiments were carried out (Table 3) for the selected fog event. In the basecase experiment of S_ARon_CRmod, the polluted aerosol particles tracked by AQC act as the source of CCN (S_) and the aerosol-radiation interaction (aerosol direct effect) is enabled in the radiation schemes (ARon). The geometric-optics approach mentioned in Section 2.3 is used to calculate the cloud optical properties of each model layer (CRmod). S_ARon_CRorig is similar to S_ARon_CRmod, except for the use of the original cloud optical property calculation (CRorig) in the NASA Goddard shortwave and longwave radiation schemes. As discussed previously, the original schemes are based on an estimate of the cloud droplet effective radius using the cloud mass and

number concentration (CRorig). The radius of cloud droplets in the original Goddard shortwave radiation scheme is constrained to the range from 4 μm to 20 μm. In the modified cloud-radiation scheme (CRmod), the size range of cloud droplets in Eq. (3) can vary between ~~the dry aerosol particle radius~~activated CCNs to 30 μm. S_AROff_CRmod has no aerosol direct effect in the radiation schemes (AROff). The comparison of S_AROff_CRmod and S_ARon_CRmod is used to estimate the aerosol direct effect in this study.

Each numerical experiment employed two domains with two-way nesting. Domain 1 (86 x 97 grid cells) had a resolution of 12 km while domain 2 (127 x 202 grid cells) had a resolution of 4 km. Domain 2 was positioned to cover the entire Central Valley of California and results from this domain are used for the subsequent analysis. All simulations used 31 vertically staggered layers based on a terrain-following pressure coordinate system. The vertical layers are stretched with a higher resolution near the surface (an average depth of ~30 m in the first model half layer). Variables other than vertical velocity and geopotential were stored in the half model levels. The time step was 60 seconds for the first domain and 20 seconds for the second domain. The physics schemes employed for the simulations included the modified Purdue Lin microphysics scheme (Chen and Sun, 2002), the NASA Goddard longwave/shortwave radiation schemes (Chou and Suarez, 1999a, 2001a), the Kain-Fritsch cumulus scheme (Kain and Fritsch, 1990; Kain, 1993) (domain 1 only), the YSU PBL scheme (Hong et al., 2006; Hong, 2010) and the Noah LSM surface scheme (Tewari et al., 2007). No cumulus scheme is used in the inner-most domain (4 km resolution). The number of cloud droplets was not considered in the convective scheme in the SOWC model. The target episode had calm winds with local fog formation in the Central Valley (not propagating in through lateral boundaries). Moreover, the event occurred in the winter season when the Convective Available Potential Energy (CAPE) was small. Therefore, the KF cumulus convective parameterization is inactive for this cases study. The meteorological

initial and boundary conditions were taken from North American Regional Reanalysis (NARR), which has a spatial resolution of 32 km and a temporal resolution of 3 hours.

The SOWC model tracked two 6D variables for aerosol/cloud properties which introduce considerable computational burden for model simulations when compared to standard WRF/Chem model simulation (with prescribed aerosol concentration). The computational cost of the SOWC model, which is proportional to the extra information that is tracked, is approximately 25 times greater than the standard WRF/Chem 3.1.1 simulation with prescribed aerosols (chem_opt = 0) or approximately 5 times greater than the standard WF/Chem 3.1.1 simulation with any chemistry option (/=0) in the current study. SOWC model simulations started at 0000 UTC 9 January (7 days prior to the start of the thick fog event) with four-dimensional data assimilation (FDDA), which nudges model fields in domain 1 to analysis including the u and v components of horizontal winds, water vapor mixing ratio, and temperature above the PBL height in all simulations. This approach provides a realistic heterogeneous aerosol distribution and low-level temperature and moisture fields at the start of the thick fog simulation. Observations from surface stations and NARR data were used for nudging during this aerosol spin-up period. Between 0000 UTC 16 January to 0000 UTC 19 January, the SOWC model integrated without FDDA (3 day free run) during which time the effects of the different model configurations were observed and is our major interested time period.

4. Model Results

4.1 Evaluation of basecase (S_ARon_CRmod) model performance

The SOWC model calculates CCN number concentrations based on the activation of aerosols (AQC). The AQC number concentration can influence the intensity of initial fog formation and spatial distribution of final fog fields, and thus AQC number concentration is

444 examined first. Figure 5 shows 72-hour averaged (from 16 to 18 January 2011) AQC number
445 concentrations in California's Central Valley that were also averaged over the first five model
446 layers for S_ARon_CRmod. Fog usually forms within the planet boundary layer (PBL),
447 which reaches to a height of approximately five model layers in winter conditions in the
448 Central Valley (450-550 m). Temporally averaged AQC concentrations are approximately
449 2×10^9 # m^{-3}, with the highest concentrations predicted in the vicinity of polluted cities (e.g.,
450 the San Francisco Bay Area, Stockton, Modesto, Sacramento, Fresno, and Bakersfield), in the
451 middle of the Central Valley, and at foothills of Sierra Nevada Mountain over the east-
452 southeastern Central Valley.

453 Figure 6 shows the comparison of simulated nitrate (NO_3^-), sulfate (SO_4^{2-}), ammonium
454 (NH_4^+) and soluble sodium (Na^+) concentrations to measured values at 6 monitoring stations
455 (see Table 2 and Fig. 4) on 18 January 2011. Simulated sulfate and soluble sodium are in
456 reasonable (>80%) agreement with measurements but nitrate and ammonium concentrations
457 were under predicted by approximately 70%. The cause for this discrepancy is unknown, but
458 one possibility is the presence of organic nitrate compounds in the atmosphere that are not
459 simulated by the model chemistry. Note that both observed and predicted nitrate
460 concentrations in the current episode are lower than the maximum concentrations observed in
461 historical extreme episodes within the San Joaquin Valley (SJV) because the current
462 stagnation event only lasted a few days while extreme events last multiple weeks. If more
463 discussion of aerosol perditions form the SOWC model is desired, we refer the reader to
464 Zhang et al. (2014) who present a comparison of predicted aerosol concentrations and
465 measured concentrations using field campaign data measured during the California Regional
466 PM_{10} / $PM_{2.5}$ Air Quality Study (CRPAQS) in December 2000 – January 2001.

467 The S_ARon_CRmod experiment reasonably reproduces the observed spatial
468 distribution and magnitude of liquid water path (LWP) compared to the data retrieved from

MODIS (Fig. 7). In particular, the model predicts LWP well over the northern portion of the Central Valley during the fog event (16 to 18 January). However, the model under-predicts LWP in the middle portion of the Central Valley, which caused the fog to dissipate earlier (late 17 January). Once the surface temperature increases in one area due to thin fog, the dissipation spreads out quickly until the fog completely evaporates. For the southern portion of the Central Valley, the fog event starts earlier (14 to 15 January) and the model reasonably predicts the onset of the event. But the simulated fog is too dense (figure not shown). In addition, the peak of the simulated fog occurs one day earlier (16 January forecast versus 17 January observed). This timing difference could be caused by the change in the microphysics processes at 0000 UTC 16 January. During the FDDA time period (before 16 January), the one-moment bulk microphysics scheme is used. After the FDDA time period, aerosols start being involved in cloud formation. High Nitrate concentrates in the SJV and enhances aerosol activation due to its high hygroscopicity. This could partially explain why the peak of the LWP occurs on 16 January. The details of aerosol chemical properties are discussed by Zhang et al. (2014).

While simulated LWP is comparable to MODIS retrievals with one day shift (Fig. 7), ~~high~~to obtain a higher COT than observed (Fig. 8b versus 8c) we expect that the model produces more small cloud droplets with a higher CCN concentration ~~and smaller cloud droplets, thus high COTs (Fig. 8), are predicted in the SOWC simulations~~, especially ~~in~~over highly polluted areas. High predicted COT results in cold surface temperature, especially in the southern portion of the Central Valley. Overall, the spatial distribution and magnitude of simulated COT also match the satellite data reasonably (Fig. 8), except for the overestimation of COT over the southeastern Central Valley (Fig. 8b and d).

Mean biases of 2-m temperature (T2), 2-m water vapor mixing ratio (Q2), and surface net downward shortwave radiative flux (NSF) over the entire Central Valley from 16 to 18

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January 2011 for S_ARon_CRmod are calculated (Fig. 9). Generally, T2 and Q2 of S_ARon_CRmod are under-predicted by 2 °C and 0.7 g kg^{-1}, respectively. The predicted time variation of T2 and Q2 biases is small in the first one and half days but increases after 1600 UTC 17 January because the predicted fog dissipated in the daytime, different from observations. Since the predicted fog dissipated, simulated NSF increased and was over-predicted by 13.9 W m^{-2}. Low simulated T2 and Q2, particularly during first one and half days, in S_ARon_CRmod are partially due to over-predictions of the fog formation (i.e., too much condensation leading to depleted water vapor), especially over the southern portion of the Central Valley. Overall, S_ARon_CRmod reasonably forecasted LWP and COT spatial pattern and intensity. S_ARon_CRmod also captured the diurnal pattern of T2 and Q2 during the fog event, but under-predicted the absolute magnitude of T2 and Q2 by 1.76 (2.22) °C and 0.56 (0.88) g kg^{-1} in the daytime (nighttime), respectively.

4.2 Source-oriented aerosol direct and indirect effects

S_ARoff_CRmod is designed to test aerosol-radiation feedback and so the comparison between S_ARoff_CRmod and S_ARon_CRmod can help quantify the aerosol direct effect in the current study. Table 4 shows that the hourly bias mean and standard deviation from 24 surface stations in the daytime and nighttime of S_ARoff_CRmod are similar to, but larger than, results from S_ARon_CRmod for T2 and Q2 at the ground. However, compared to S_ARon_CRmod, the smaller cold bias from S_ARoff_CRmod is consistent with its larger net downward shortwave radiative flux (NSF) shown in Tables 4 and 5. Table 5 shows that the average NSF within the entire Central Valley from S_ARoff_CRmod is higher than S_ARon_CRmod by 3.7 W m^{-2}, which means that the shortwave energy flux that reached the ground was reduced by ~3.7 W m^{-2} due to aerosol radiative forcing in this case study. The maximum increases of T2 and NSF by the aerosol direct effect occurred on 17 January 2011 (Fig. 9). Table 5 also shows the mean value of cloud liquid water ~~mixing ratio~~, cloud droplet

number, surface skin temperature, latent heat flux and sensible heat flux over the Central Valley during 16 to 18 January 2011. Cloud liquid water ~~mixing ratio~~ and cloud droplet number were averaged within the first five model layers. The aerosol direct effect leads to increases in the cloud water mass and cloud droplet number by 3.3% and 4.5%, respectively, due to reductions in skin temperature (0.1 K) and net shortwave flux (3.7 W m^{-2}).

The modified radiation schemes for cloud optical properties in the S_ARon_CRmod experiment do not have significant feedback on spatially and temporally averaged cloud water mass ~~and cloud droplet number~~ (i.e., compared to S_ARon_CRorig) as shown in Table 5. ~~Theoretically~~However, the modified cloud-radiation interaction (i.e., geometric-optics method) used in the COT calculations (S_ARon_CRmod) ~~can predict~~predicts slightly higher COT, which leads to slightly lower net shortwave flux and surface skin temperature, especially in the polluted area. The higher COT predictions are likely caused by differences in the size range of cloud droplets and refractive indexes of cloud water with/without chemical composition in the calculation of cloud radiative properties. ~~As mentioned above, the~~In the original radiation scheme, the cloud optical thickness (COT) is a function of cloud water path (CWP) and an effective radius ($4\ \mu m \leq r_e \leq 20\ \mu m$), which is derived from the water mass and the total droplet number assuming a uniform size of cloud droplets ~~in the original Goddard shortwave radiation scheme is constrained to the range from 4 μm to 20 μm, while in our~~ :

$$\tau_{orig}(\lambda) = CWP \times (-6.59 \times 10^{-3} + \frac{1.65}{r_e}). \quad (6)$$

In the modified radiation scheme~~, the cloud droplets~~ in S_ARon_CRmod, the COT is calculated based on simulated cloud droplet size, number, and chemical composition of each bin and source (Eq. 3). In addition, the formula of COT, single scattering albedo and asymmetry factor in the modified radiation scheme are ~~allowed to range~~all modified. With a similar Q_c, although Q_n in ~~size~~ S_ARon_CRorig is higher than that in S_ARon_CRmod, the

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COT is slightly higher in S_ARon_CRmod due to different formulas used in the calculation of cloud-radiation interaction. The small difference of COT between ~~the dry aerosol particle radius to 30 μm. The~~ these two experiments in fact indicates that the parameterization of ~~cloud optical thickness~~ COT in the original ~~Goddard radiation scheme assumes that cloud droplets are pure water. The modified~~ radiation scheme ~~recognizes the chemical species in the cloud water and considers these species when calculating the cloud droplet index of refraction. However, in this case study the results of these two experiments (i.e., S_ARon_CRmod and S_ARon_CRorig) were very similar.~~ provides a reasonable result compared to the explicit COT calculation.

Because the meteorological conditions of the fog event are calm and stable, the cloud microphysics processes are fairly slow and simple (no rain produced in this case). Although S_ARon_CRorig had slightly higher cloud droplet number concentrations, the modified calculation of the cloud optical properties in S_ARon_CRmod gave a similar cloud amount and net shortwave radiation flux reaching the surface, which produced nearly identical feedbacks to meteorology in both experiments (Table 5).

4.3 Internal mixture versus source-oriented aerosols

The mixing state of chemical components among the atmospheric aerosol particles can potentially play an important role in fog formation. The activation of aerosol particles into cloud droplets depends on the critical ~~super-saturation~~supersaturation which in turn depends on particle composition. According to the Köhler equation, increased concentrations of solutes will decrease the critical ~~super-saturation~~supersaturation required to activate a particle into a CCN. As mentioned earlier, hydrophobic particles (i.e. black carbon) will more easily serve as CCN once they are coated with hygroscopic material (i.e. sulfate). Increased concentrations of solutes can potentially modify the frequency and severity of fog events in polluted air. In this section, we compare results from S_ARon_CRmod (source-oriented (i.e.,

aging-process-included) experiment) and I_ARon_CRmod (internally mixed experiment) to investigate the activation change and further meteorological responses between internally mixed and source-oriented aerosols. The internally mixed experiment is conducted by lumping all sources together (i.e., AQC source dimension collapsed to one producing a 5D AQC variable).

It is likely that the ratio of CCN concentration (N_{CCN}) to total aerosol concentration (N_{CN}) will be different for each of the five source types tracked in S_ARon_CRmod since the CCN activation depends on the chemical composition and size of the particles. The highest ratio of N_{CCN}/N_{CN} for S_ARon_CRmod and I_ARon_CRmod is located in the southern Central Valley (Fig. 10) due to higher moisture from the atmospheric river resulting in greater aerosols activation to CCNs and smaller residual aerosol number concentration (see Fig. 5). Over the Central Valley during 16 to 18 January 2011, the ratio of N_{CCN}/N_{CN} for each source type is 12.63%, 15.60%, 14.89%, 16.80% and 20.21% for wood smoke, gasoline, diesel, meat cooking, and others, respectively (averaged within the first five model layers). Wood smoke is typically a major source of aerosol (~38%) in California's Central Valley during winter stagnation events (see Table 6) and the organic carbon in wood smoke is water-soluble (Dusek et al., 2011) which allows these particles to activate more easily than insoluble particles. However, the majority of the wood smoke particles are located in the smallest size bin, so the ratio of N_{CCN}/N_{CN} for wood smoke is comparable with that of hydrophobic diesel. The source type of "others", which has the highest ratio of N_{CCN}/N_{CN}, is dominated by larger dust particles coated with secondary components such as nitrate and are easier to activate, in contrast to the smaller combustion particles emitted from other tracked sources.

The comparison of the average ratio of N_{CCN}/N_{CN} from the first five model layers between S_ARon_CRmod and I_ARon_CRmod is shown on Fig. 10. The spatial patterns

produced by both experiments are similar but I_ARon_CRmod has a higher N_{CCN}/N_{CN} ratio, in particular over the northern two thirds of the Central Valley. The largest differences between N_{CCN}/N_{CN} predicted by S_ARon_CRmod and I_ARon_CRmod occur in regions with large emissions of wood smoke (figure not shown). The ratio of N_{CCN}/N_{CN} for both experiments can reach >30% but the highest N_{CCN}/N_{CN} ratio occurs in relatively less polluted regions. The spatially averaged ratio of N_{CCN}/N_{CN} is 16.65% for S_ARon_CRmod and 27.49% for I_ARon_CRmod within the Central Valley over the period of 16 to 18 January. The CCN concentrations and N_{CCN}/N_{CN} ratios between internally mixed and source-oriented experiments at different ~~super-saturations~~supersaturations were calculated to better understand this result. Figure 11a shows the 72-hour averaged CCN concentration at ~~super-saturations~~supersaturations of 0.02%, 0.05%, 0.1%, 0.2% and 0.5% and total AQC concentration averaged within the first five model layers. Figure 11b presents corresponding N_{CCN}/N_{CN} ratios at 5 different ~~super-saturations.~~supersaturations. When the ~~super-saturation~~supersaturation is less than or equal to 0.2%, the N_{CCN}/N_{CN} ratio predicted from S_ARon_CRmod is comparable or even slightly higher than that predicted from I_ARon_CRmod. In the S_ARon_CRmod tests, 56% of the particles tracked in the AQC variable (mainly in size bins 2-8) are activated as CCN. When the ~~super-saturation~~supersaturation is close to 0.5%, the N_{CCN}/N_{CN} ratio from I_ARon_CRmod can be 15% higher than that of S_ARon_CRmod. Most particles tracked in the AQC size bin 1 can activate in the internally mixed experiment; however, in the source-oriented experiment only particles ~~in AQC size bin 1 associated with wood smoke and "others" sources activate due to the relatively hydrophobic nature of particles associated with other sources (Table 6). Cubison et al. (2008)~~emitted in AQC size bin 1 from wood smoke and "others sources" are sufficiently hygroscopic to activate. The remaining sources are dominated by hydrophobic compounds (such as elemental carbon) that do not activate under the study conditions (Table

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6). Cubison et al. (2008) analyzed observational CCN and CN data in 2005 from a field campaign in California and found that the average ratio of N_{CCN}/N_{CN} was 18% for a ~~super-saturation~~supersaturation value of 0.5%, but their predicted N_{CCN}/N_{CN} ratio based on the internal mixture assumptions could reach to more than 50%. In the source-oriented SOWC model, ~~super-saturation~~supersaturation values are typically ~0.2-0.3% with maximum value of 0.5% in some areas. The estimated ratio of N_{CCN}/N_{CN} in the source-oriented model is comparable with observations in Cubison et al. (2008), especially in polluted areas. The temporal variations of mean bias of 2-m temperature (T2), 2-m water vapor mixing ratio (Q2), and surface net downward shortwave radiative flux (NSF) between internal versus external aerosol mixture states (I_ARon_CRmod versus S_ARon_CRmod) are similar ~~untill~~until 2000 UTC 17 January. After late 17 January, the bias differences between two experiments are more apparent in the daytime than in the nighttime (Fig. 9 and Table 4). Compared to I_ARon_CRmod, S_ARon_CRmod reduced bias in T2 by 0.25 K in the daytime but had higher bias in NSF. S_ARon_CRmod did predict improved values of Q2. Based on Fig. 9, we know that the source-oriented and internal aerosol mixing states mainly cause differences in surface temperature in the daytime. Figures 12a and b illustrate the relative change ((internally mixed – source-oriented)/source-oriented * 100%) of averaged (16 - 18 January 2011) cloud liquid water~~ mixing ratio~~ and cloud droplet number, respectively, during the daytime. I_ARon_CRmod predicts cloud liquid water~~ mixing ratios~~ that are 40% higher than values predicted by S_ARon_CRmod over the northern Central Valley (Fig. 12a). The largest relative change in predicted cloud water concentration also occurs in the northern Central Valley near the mountains where fogs are initiated by drainage flow. I_ARon_CRmod predicts higher cloud droplet number (Fig. 12b), with the largest relative increases ~~(~~~(50~~–~~~60%) once again observed in areas near mountains and highly polluted regions with more modest changes of 20~30% over remote regions. Internally

644 mixed aerosols reduce the critical saturation ratio for particles by artificially mixing
645 hygroscopic and hydrophobic components that in turn allows particles to activate more
646 easily.

647 The internally mixed experiment (I_ARon_CRmod) predicts lower daytime averaged
648 surface skin temperature and net downward shortwave flux at ground (Fig. 12c and d)
649 corresponding to the areas with higher cloud liquid water ~~mixing ratio~~ and cloud droplet
650 concentrations (Fig. 12a and b). This result is expected since higher cloud liquid water
651 ~~mixing ratio~~ and cloud droplet concentration will reduce the solar radiation flux on the
652 surface. The reduction of surface skin temperature in the internal mixed experiment is
653 proportional to the change of the net shortwave radiation. Figure 13 shows that the area
654 average of latent heat flux (LH) and sensible heat flux (SH) over the Central Valley in
655 S_ARon_CRmod and the average difference of internally mixed and source-oriented
656 experiments. Higher cloud amount and lower surface temperature are predicted in the
657 internally mixed experiment leading to reduced LH and SH fluxes at ground level compared
658 to the source-oriented experiment. The difference between internally mixed and source-
659 oriented predictions for LH and SH reached 3 W m^{-2} and 5 W m^{-2}, respectively, at noon local
660 time (2200 UTC 17 January).

661 Table 7 shows hourly mean bias and root-mean-square-difference between internally
662 mixed (I_ARon_CRmod) and source-oriented (S_ARon_CRmod) experiments for six
663 variables within the Central Valley during 16 to 18 January 2011. The mean bias between
664 these two experiments is 1.19×10^{-2} (g m^{-3}) for cloud liquid water ~~mixing ratio~~ and 6.24×10^{7}
665 (# m^{-3}) for cloud droplet number. The direction of these trends is expected since internally
666 mixed aerosols are easier to activate as CCN. The mean bias between internally mixed and
667 source-oriented experiments is -0.15 (K) for surface skin temperature and -6.02 (W m^{-2}) for
668 net shortwave flux. The mean bias of LH and SH is -0.61 and -0.36 (W m^{-2}), respectively.

The root-mean-square-difference between these two experiments is large for each variable, meaning that the difference varies strongly with location (see Fig. 12).

In summary, compared to S_ARon_CRmod, I_ARon_CRmod has a higher CCN / cloud droplet number concentration because internally mixed aerosols can instantaneously contain hydroscopic material (e.g. sulfate) through artificial mixing, which decreases the critical supersaturation requirement for a particle to activate into a CCN, leading to higher cloud number concentration and optical thickness. Thicker fog in I_ARon_CRmod reduces the amount of shortwave radiation reaching the surface, resulting in a lower surface temperature. A lower surface temperature can have a positive feedback on cloud lifetime (i.e., a longer cloud lifetime), which further reduces shortwave radiation reaching the surface when compared to S_ARon_CRmod. Hence, the aerosols that include aging processes can delay CCN activation and produce fewer cloud droplets and less fog, which in turn will modify the energy budget near the surface.

5. Summary and discussion

A warm cloud-aerosol interaction module was implemented into the source-oriented Weather Research and Forecasting model with Chemistry (SOWC) to study the aerosol-cloud-radiation interactions during fog simulations. The source-oriented mixture of aerosols is used to explicitly simulate particle aging processes in the atmosphere rather than instantaneously combining particles into an internal mixture. The SOWC model was used to simulate a fog event in California's Central Valley in January 2011 with seven days of FDDA nudging and three days of free run. Fog formation occurred when high moisture content from an Atmospheric River was advected into the Central Valley and cold drainage flows occurred into the valley at night. The initial tests used 5 emissions sources (wood smoke, gasoline, diesel, meat cooking, and others) with particles from each source consisting of 38

chemical species and 8 size bins, spanning a diameter range from 0.01 to 10 microns. The highest model spatial resolution was 4 km.

Four numerical experiments were conducted to test model performance, meteorological feedbacks from internal and source-oriented aerosols, and the impact of aerosol-cloud-radiation interaction on fog formation. Compared to observations, the SOWC model reasonably predicted fog spatial distribution and duration and environmental meteorological feedbacks. However, the model over-predicted liquid water path and cloud optical thickness, which resulted in cold surface temperature bias. The inclusion of aerosol-radiation interaction reduced net downward shortwave radiative flux by an average of 3.7 W m^{-2} and daytime surface temperature by 0.1 K. Results that used different treatments for aerosol mixing states were compared, and the important ~~findings~~results are: 1) the fraction of N_{CCN}/N_{CN} at a supersaturation of 0.5% in the Central Valley decreased from 94% in the internal mixture model to 80% in the source-oriented mixture model; 2) due to a smaller number of the CCN concentration in the source-oriented mixture model than in the internal mixture model, cloud liquid water ~~mixing ratio~~ and cloud droplet number decreased 5% and 15%, respectively; and 3) compared to observations, the source-oriented mixture model reduced the cold bias for surface temperature by 0.25 K in the daytime relative to the internal mixture model. The source-oriented mixture representation of particles also provided more reasonable predictions for cloud droplet number and cloud water mass versus observations due to different activation properties than the internal mixture representation of particles. The internal mixture model predicted greater activation of CCN than the source-oriented model due to artificial coating of hydrophobic particles with hygroscopic components.

The SOWC model in this study explicitly calculates primary particle aging over a regional scale for fog formation prediction with two-moment microphysics scheme and aerosol-cloud-radiation interactions. The SOWC model should be a useful tool to study

aerosol-cloud-radiation interactions. ~~We are now conducting more numerical studies on different weather systems to explore the full range of responses~~Note that the current results are based on a Tule fog case study in the Central Valley of California. Additional fog case studies under different weather conditions in other regions of the world are required to draw conclusions at those locations.

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Captions of Tables

Table 1. Chemical species that are carried in the AQC/CLDAQC "species" dimension. All species are in concentrations (μg m^{-3}) except for the last two elements (i.e., 39 and 40), which carry the number concentration (# m^{-3}) and radius (m).

Table 2. California Ambient Air Quality Data (CAAQD) station information.

Table 3. Numerical experiment designs for this study.

Table 4. Hourly bias mean and standard deviation (std) in day time and night time of 2-m temperature (T2, °C), water vapor mixing ratio (Q2, g kg-air^{-1}), and net downward shortwave radiative flux (NSF, W m^{-2}) between all experiments and observation from 16 to 18 January 2011. T2 and Q2 are calculated using 24 surface stations and NSF is calculated using 42 CIMIS stations shown in Fig. 4.

Table 5. Mean values of cloud liquid water ~~mixing ratio~~ (Q_c), cloud droplet number (Q_n), surface skin temperature (SKT), net shortwave flux (NSF), latent heat flux (LH) and sensible heat flux (SH) for four experiments over the entire Central Valley during 16 to 18 January 2011.

Table 6. ~~Ratio~~The ratio of AQC number concentration for each bin/source to the total number concentration. The numbers are averaged within the first five model layers during 16 to 18 January 2011.

Table 7. Hourly bias mean and root-mean-square-difference of cloud liquid water ~~mixing ratio~~ (Q_c), cloud droplet number (Q_n), surface skin temperature (SKT), net shortwave flux (NSF), latent heat flux (LH) and sensible heat flux (SH) between internally mixed (I_ARon_CRmod) and source-oriented (S_ARon_CRmod) experiments (internally mixed – source-oriented) during 16 to 18 January 2011.

Captions of Figures

Figure 1. Cloud physics processes that are involved with cloud particles in the SOWC model with a 6D aerosol variable (AQC) and a 6D cloud variable (CLDAQC) included. Black solid arrow and grey dashed arrow indicate the source and the sink processes of cloud water and 6D CLDAQC, as well as 6D AQC, respectively.

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Figure 8. Same as Figure 5 but cloud optical thickness (COT) (dimensionless).

Figure 9. Mean bias variation of (a) 2-m temperature (T2), (b) 2-m water vapor mixing ratio (Q2), and (c) surface net downward shortwave radiative flux (NSF) between observations and model simulation from 16 to 18 January 2011 for S_ARon_CRmod (blue lines), S_AROff_CRmod (purple lines) and I_ARon_CRmod (red lines) experiments.

Figure 10. N_{CCN}/N_{CN} ratio for (a) S_ARon_CRmod (source-oriented experiment) and (b) I_ARon_CRmod (internally mixed experiment) averaged within the first five model layers. The ratio is hourly average during 16 to 18 January 2011. Contours are terrain heights in m.

Figure 11. (a) 72-hour averaged CCN concentration at supersaturation of 0.02%, 0.05%, 0.1%, 0.2%, 0.5% and total AQC concentration with units in # cm^{-3}. (b) N_{CCN}/N_{CN} ratio at 5 corresponding supersaturation. Dark gray is source-oriented experiment and light gray represents internally mixed experiment. Results are average values using data within the first five model layers.

Figure 12. Relative change ((internally mixed – source-oriented)/source-oriented * 100%) in 72-hour averaged predictions during 16 to 18 January 2011 for (a) the ratio of cloud liquid water ~~mixing ratio~~, (b) cloud droplet number, (c) surface skin temperature and (d) net shortwave radiation. (a) and (b) are average values using data within the first five model layers. Contours are terrain heights in m.

Figure 13. Area average of latent heat flux (LH) and sensible heat flux (SH) over the Central

1008 Valley in S_ARon_CRmod and the average difference between I_ARon_CRmod and
1009 S_ARon_CRmod from 0800 UTC 17 January (00 Z local time) to 0700 UTC 18 January
1010 (23 Z local time).

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1012

1013 Table 1. Chemical species that are carried in the AQC/CLDAQC "species" dimension. All
1014 species are in concentrations (μg m^{-3}) except for the last two elements (i.e., 39 and 40), which
1015 carry the number concentration (# m^{-3}) and radius (m).

	Chemical species		Chemical species
1	EC	21	SOA from lumped Alkane 1
2	OC	22	SOA from lumped Alkane 2
3	NA	23	SOA from lumped Aromatic 1
4	CL	24	SOA from lumped Aromatic 2
5	N3	25	SOA from lumped Aromatic 1
6	S6	26	SOA from lumped Aromatic 2
7	N5	27	SOA from lumped Aromatic 1
8	Other	28	SOA from lumped Aromatic 2
9	Metal	29	SOA from lumped Alkene 1
10	Unknown	30	SOA from lumped Alkene 2
11	CU1	31	SOA from lumped Alpha Pinene 1
12	CU2	32	SOA from lumped Alpha Pinene 2
13	MN2	33	SOA from lumped Beta Pinene 1
14	MN3	34	SOA from lumped Beta Pinene 2
15	FE2	35	SOA from lumped Toluene 1
16	FE3	36	SOA from lumped Toluene 2
17	S4	37	Hydrogen Ion
18	Air (hollow sea salt particles)	38	Water
19	NO3	39	Number Concentration
20	Non-explicit SOA	40	Particle Mean Volume Radius

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Table 2. California Ambient Air Quality Data (CAAQD) station information

Station ID	Station name	Longitude (º)	Latitude (º)
1	San Jose-Jackson Street	-121.89	37.35
2	Bakersfield-5558 Cal. Avenue	-119.06	35.36
3	Fresno-1st Street	-119.77	36.78
4	Modesto-14th Street	-120.99	37.64
5	Visalia-N Church Street	-119.29	36.33
6	Sacramento-T Street	-121.49	38.57

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1026 Table 3. Numerical experiment designs for this study.

Experiments	Description
S_ARon_CRmod	Source-Oriented aerosols with aerosol direct effect calculation on and modified cloud radiation parameterization
S_ARon_CRorig	Source-Oriented aerosols with aerosol direct effect calculation on and original cloud radiation parameterization
S_ARoff_CRmod	Source-Oriented aerosols with aerosol direct effect calculation off and modified cloud radiation parameterization
I_ARon_CRmod	Internal mixing aerosols with aerosol direct effect calculation on and modified cloud radiation parameterization

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Table 4. Hourly bias mean and standard deviation (std) in day time and night time of 2-m temperature (T2, °C), water vapor mixing ratio (Q2, g kg-air^{-1}), and net downward shortwave radiative flux (NSF, W m^{-2}) between all experiments and observation from 16 to 18 January 2011. T2 and Q2 are calculated using 24 surface stations and NSF is calculated using 42 CIMIS stations shown in Fig. 4.

	S_ARon_CRmod		S_ARon_CRorig		S_AROff_CRmod		I_ARon_CRmod	
Daytime	Bias mean	std	Bias mean	std	Bias mean	std	Bias mean	std
T2	-1.76	1.27	-1.72	1.32	-1.63	1.33	-2.01	1.09
Q2	-0.56	0.34	-0.56	0.36	-0.54	0.35	-0.57	0.32
NSF	13.91	53.18	14.40	58.00	18.81	58.78	8.68	50.03
Nighttime	Bias mean	std	Bias mean	std	Bias mean	std	Bias mean	std
T2	-2.22	0.92	-2.21	0.95	-2.19	0.93	-2.30	0.87
Q2	-0.88	0.41	-0.87	0.42	-0.88	0.42	-0.89	0.41
NSF	/	/	/	/	/	/	/	/

Table 5. Mean values of cloud liquid water ~~mixing ratio~~ (Q_c), cloud droplet number (Q_n), surface skin temperature (SKT), net shortwave flux (NSF), latent heat flux (LH) and sensible heat flux (SH) for four experiments over the entire Central Valley during 16 to 18 January 2011.

	S_ARon_CRmod	S_ARon_CRorig	S_ARoff_CRmod	I_ARon_CRmod
Q_c^* (g m^{-3})	0.220	0.221	0.213	0.231
Q_n^* (# m^{-3})	3.94×10^8	4.18×10^8	3.77×10^8	4.57×10^8
SKT (K)	281.305	281.30	281.404	281.151
NSF** (W m^{-2})	130.56	131.02	134.24	124.54
LH (W m^{-2})	9.01	9.02	9.36	8.40
SH (W m^{-2})	4.91	4.55	5.27	4.54
COT (unitless)	25.56	25.15	24.49	28.62

* Averaged within the first five model layers

** Averaged only in the daytime

Table 6. Ratio of AQC number concentration for each bin/source to the total number concentration. The numbers are averaged within the first five model layers during 16 to 18 January 2011.

	Wood smoke	Gasoline	Diesel	Meat cooking	Others	Source-oriented	Internal
Bin1	28.92%	1.00%	4.25%	0.84%	10.39%	45.40%	48.89%
Bin2	9.12%	0.38%	1.48%	0.60%	38.64%	50.22%	46.74%
Bin3	0.19%	0.01%	0.03%	0.02%	3.03%	3.28%	3.26%
Bin4	0.00%	0.00%	0.00%	0.00%	0.17%	0.18%	0.21%
Bin5	0.00%	0.00%	0.00%	0.00%	0.02%	0.02%	0.02%
Bin6	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Bin7	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Bin8	0.00%	0.00%	0.00%	0.00%	0.91%	0.91%	0.88%

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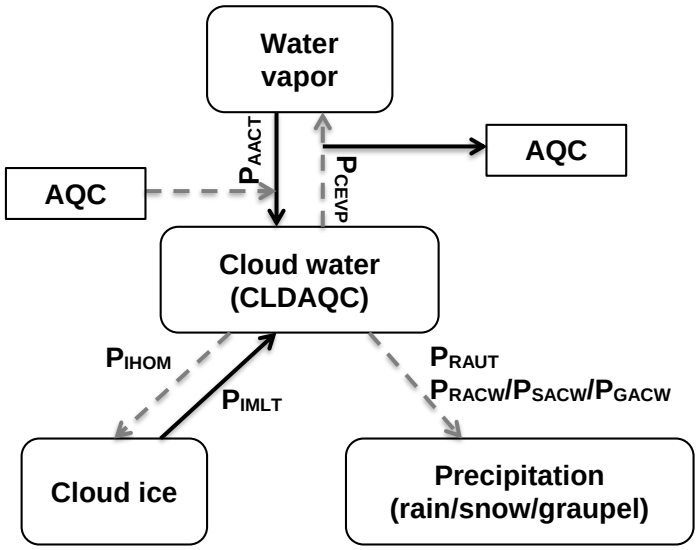
Table 7. Hourly bias mean and root-mean-square-difference of cloud liquid water ~~mixing ratio~~ (Q_c), cloud droplet number (Q_n), surface skin temperature (SKT), net shortwave flux (NSF), latent heat flux (LH) and sensible heat flux (SH) between internally mixed (I_ARon_CRmod) and source-oriented (S_ARon_CRmod) experiments (internally mixed – source-oriented) during 16 to 18 January 2011.

	Bias mean	**Root-mean-square-difference**
Q_c^* (g m^{-3})	1.19×10^{-2}	4.16×10^{-2}
Q_n^* (# m^{-3})	6.24×10^{7}	2.64×10^{8}
SKT (K)	-0.15	0.57
NSF (W m^{-2})	-6.02	13.30
LH (W m^{-2})	-0.61	2.75
SH (W m^{-2})	-0.36	5.24

* Averaged within the first five model layers



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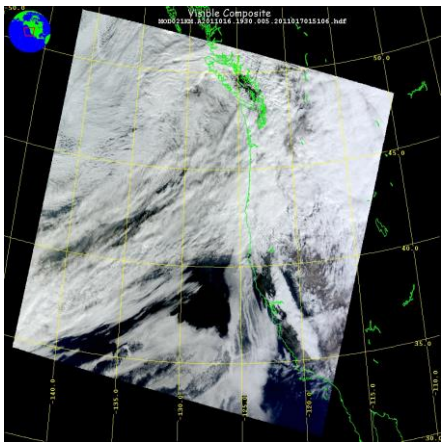
(a)



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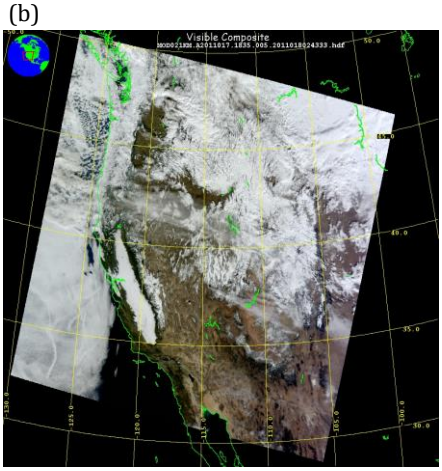

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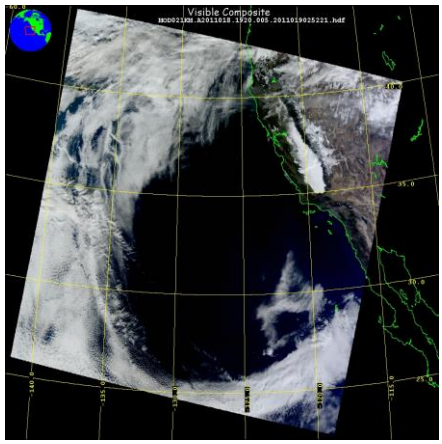

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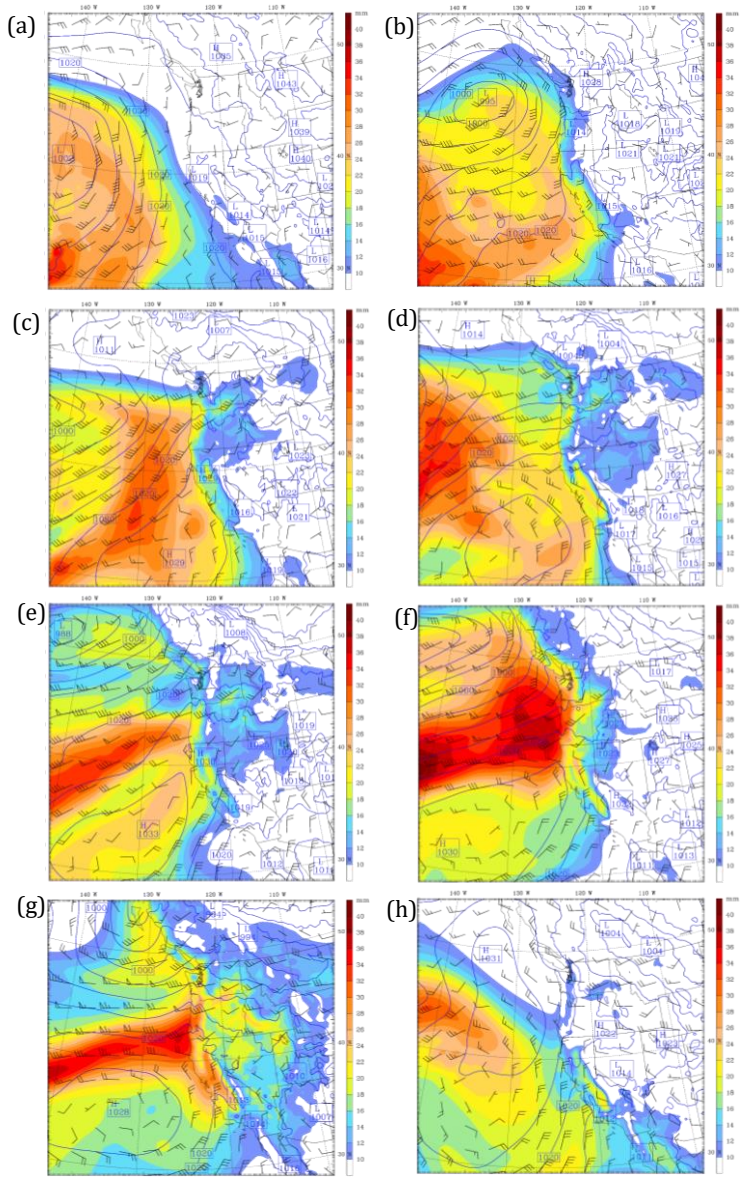

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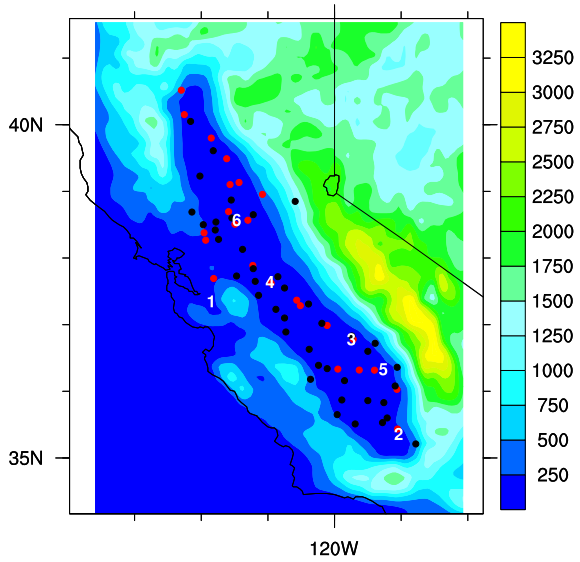

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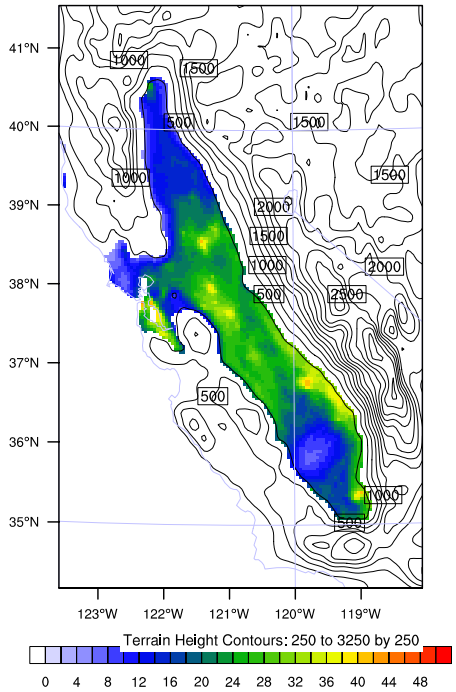

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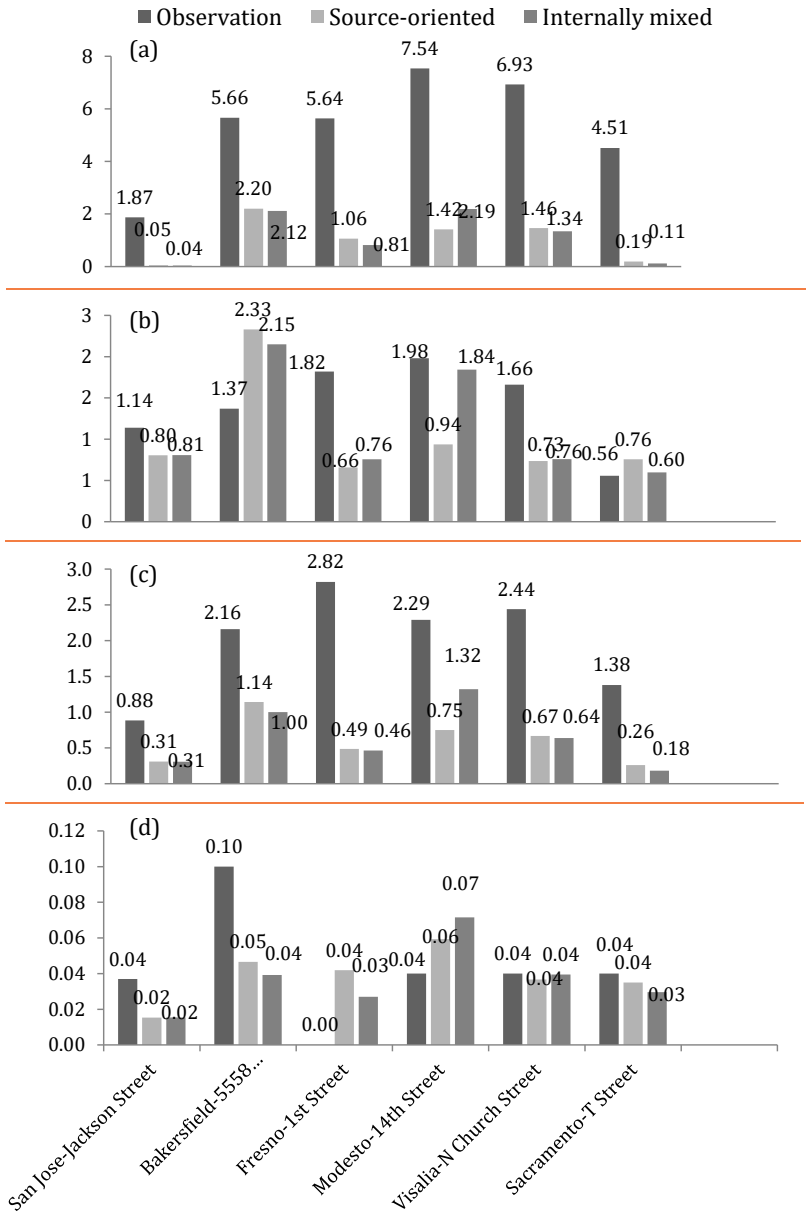

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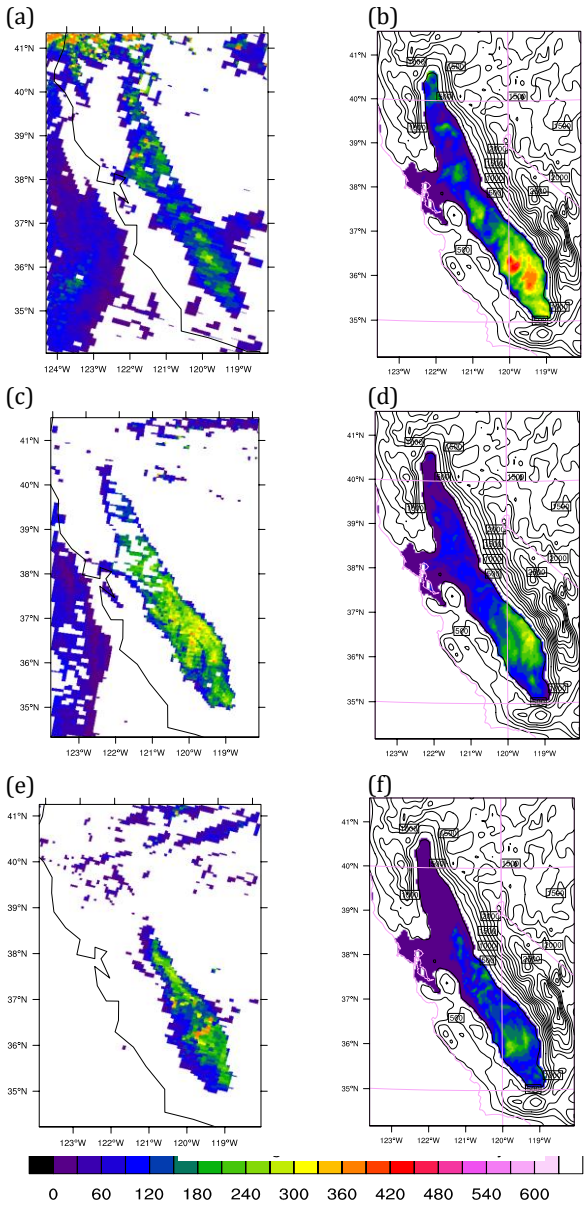

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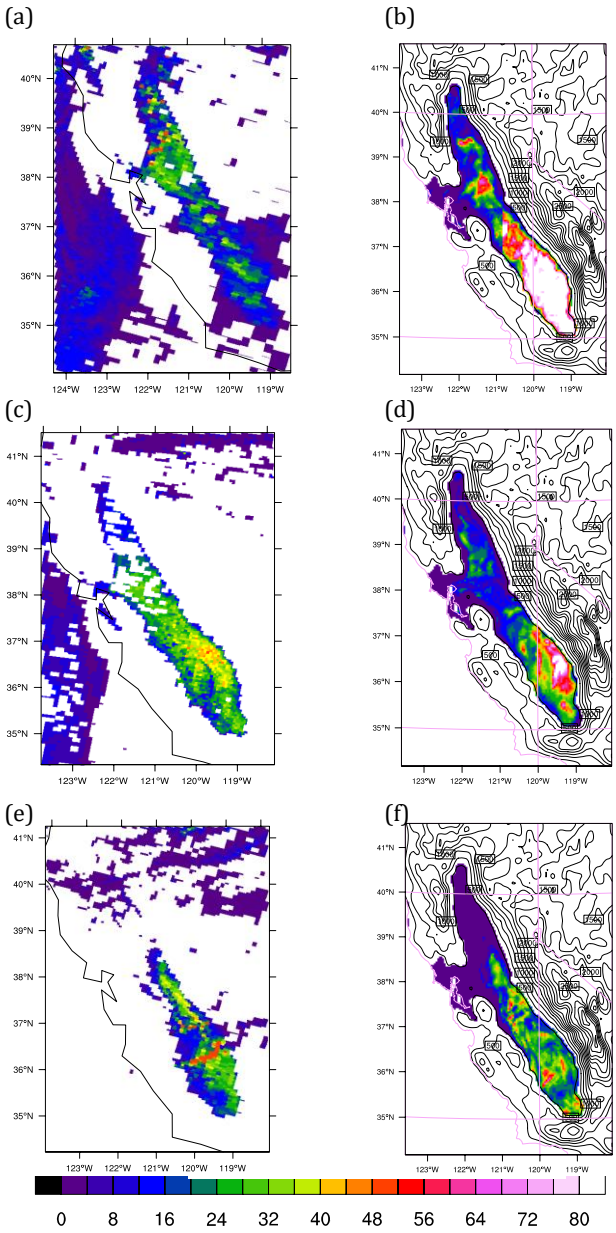

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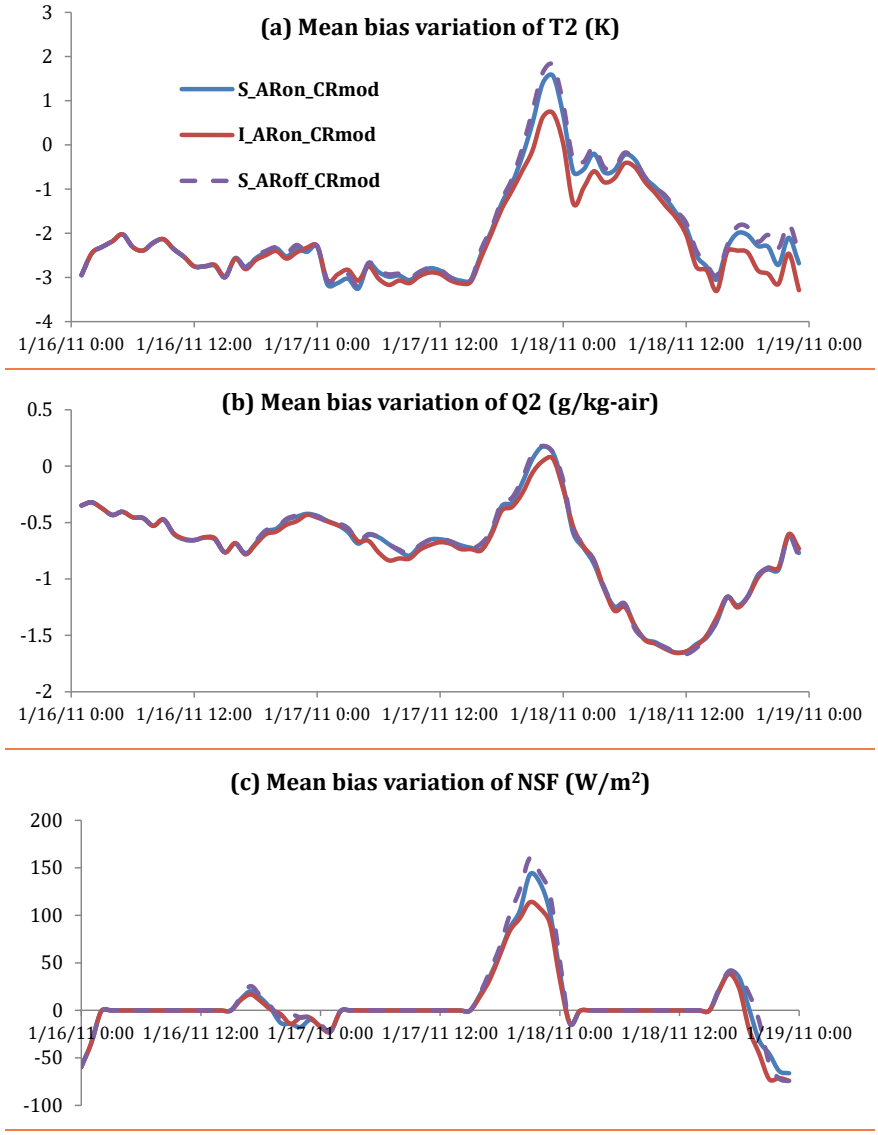

Figure 9. Mean bias variation of (a) 2-m temperature (T2), (b) 2-m water vapor mixing ratio (Q2), and (c) surface net downward shortwave radiative flux (NSF) between observations and model simulation from 16 to 18 January 2011 for S_ARon_CRmod (blue lines), S_ARoff_CRmod (purple lines) and I_ARon_CRmod (red lines) experiments.

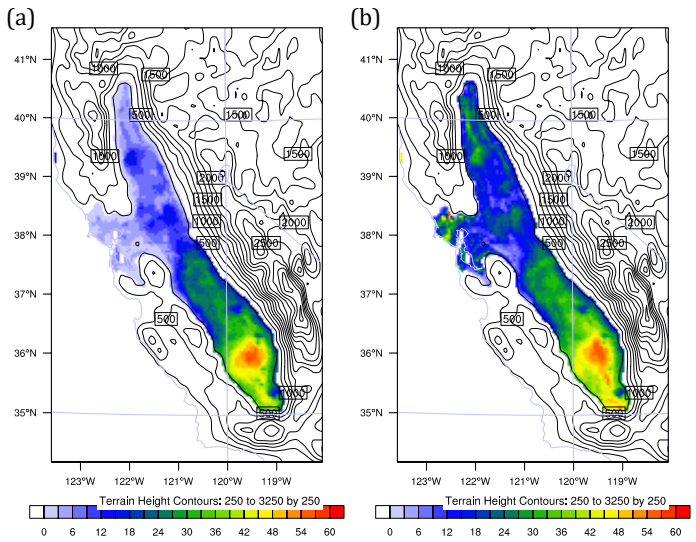

Figure 10. N_{CCN}/N_{CN} ratio for (a) S_ARon_CRmod (source-oriented experiment) and (b) I_ARon_CRmod (internally mixed experiment) averaged within the first five model layers. The ratio is hourly average during 16 to 18 January 2011. Contours are terrain heights in m.

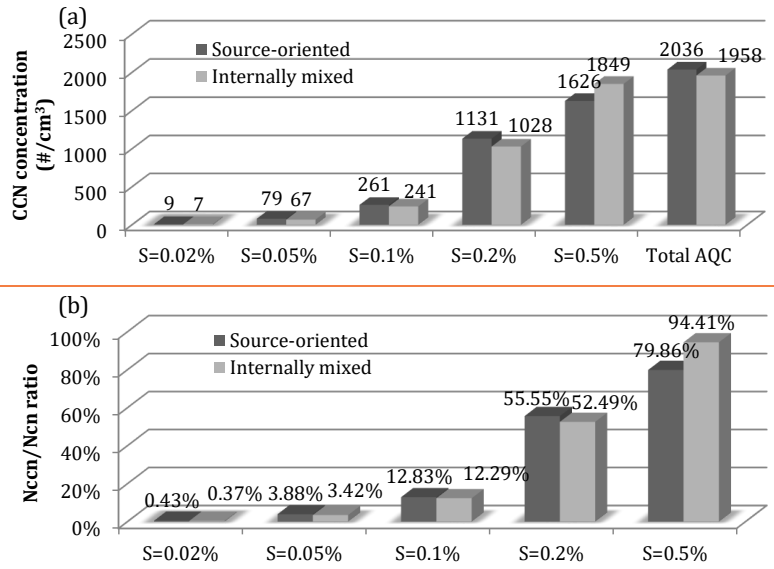

Figure 11. (a) 72-hour averaged CCN concentration at supersaturation of 0.02%, 0.05%, 0.1%, 0.2%, 0.5% and total AQC concentration with units in # cm^{-3}. (b) N_{CCN}/N_{CN} ratio at 5 corresponding supersaturation. Dark gray is source-oriented experiment and light gray represents internally mixed experiment. Results are average values using data within the first five model layers.

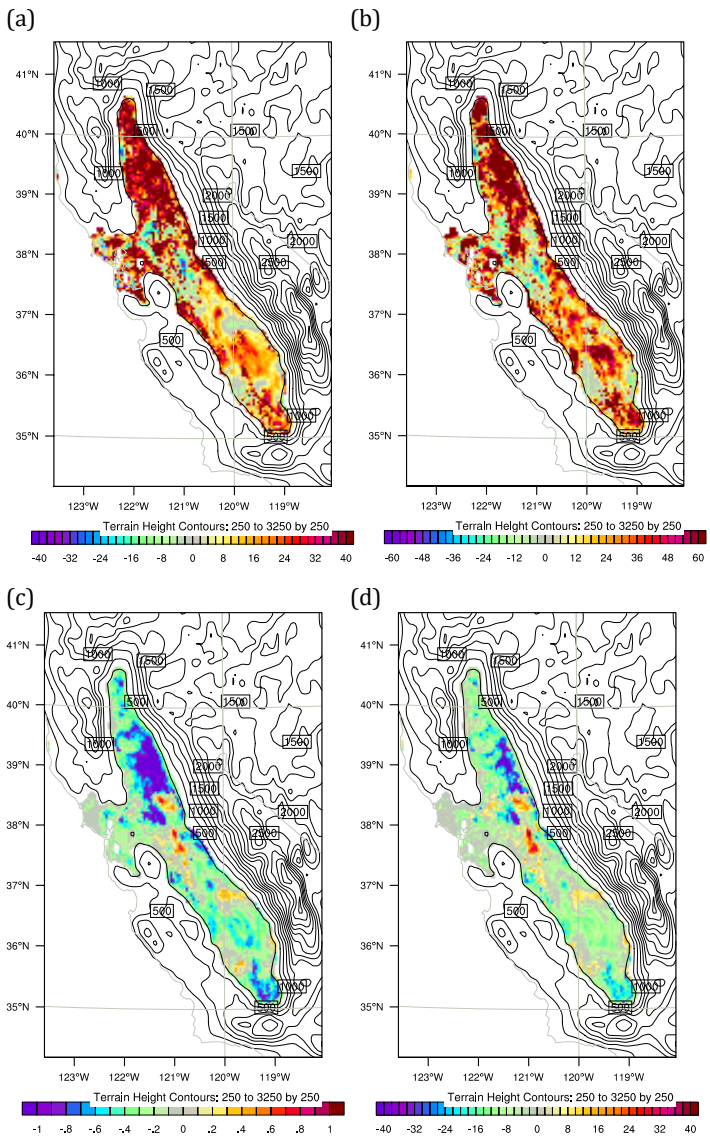

Figure 12. Relative change ((internally mixed – source-oriented)/source-oriented * 100%) in the daytime averaged predictions during 16 to 18 January 2011 for (a) the ratio of cloud liquid water ~~mixing ratio~~, (b) cloud droplet number and absolute difference (internally mixed – source-oriented) in (c) surface skin temperature (K) and (d) net shortwave radiation (W m^{-2}). (a) and (b) are average values using data within the first five model layers. Contours are terrain heights in m.

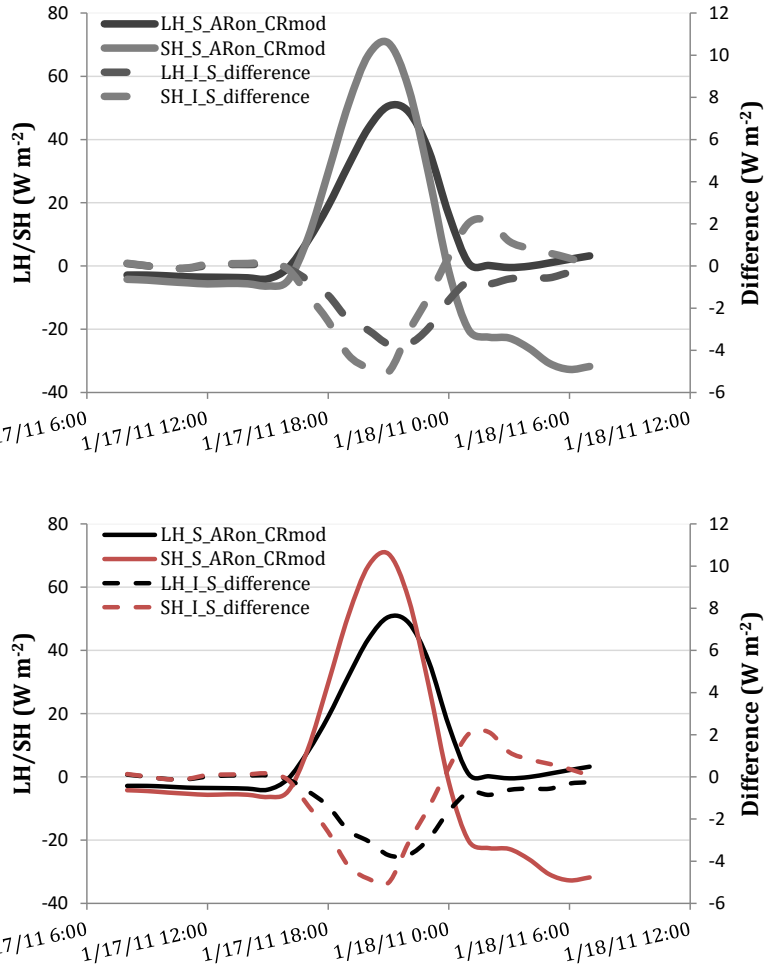

Figure 13. Area average of latent heat flux (LH) and sensible heat flux (SH) over the Central Valley in S_ARon_CRmod and the average difference between I_ARon_CRmod and S_ARon_CRmod from 0800 UTC 17 January (00 Z local time) to 0700 UTC 18 January (23 Z local time).