Anonymous Referee #2

This paper well describes the impacts of source-oriented aerosols and aerosol-cloud interaction on fog formation by implementing the modified cloud microphysics and radiation schemes into the source-oriented Weather Research and Forecasting chemistry model (SOWC). Here are some major and specific comments, which need to be considered before the publication.

The authors appreciate the reviwer'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.*

1. The authors noted in section3 that the computational cost of the SOWC model simulation is 25 times higher than that of the standard WRF/Chem simulation. Known that the SOWC model is computationally very expensive, how can authors conclude that the SOWC model should be a useful public model to predict effects of climate change on the hydrological cycle and energy budget?

Reply: The standard WRF/Chem simulation we mentioned in the manuscript is the WRF/Chem model with prescribed aerosols (chem_opt = 0), which does not include any chemistry processes. In general, the computational cost of WRF/Chem with any chemistry option (/=0) is about 5 times of that with chem_opt = 0 in the released WRF/Chem 3.1.1 version. Thus, the computational time of SOWC is about 5 times of WRF/Chem with any chemistry option (/=0). Although the SOWC model still has a higher computational demand and is probably not feasible for all users, it should be a useful tool to users who are able to access super computers or computer clusters to conduct research relevant to aerosol-cloud-radiation interactions. With rapidly growing CPU efficiency and computing resources it seems a natural step to move on to the next stage of research for pursuing a more comprehensive method with fewer assumptions, like the source-oriented method, to study aerosol-cloud-radiation interactions.

We appreciate the reviewer's comment and have modified "the SOWC model should be a useful public model to study aerosol-cloud-radiation interactions and to predict the effects of climate change on the hydrological cycle and energy budget." to **"the SOWC model should be a useful tool to study aerosol-cloud-radiation interactions"** in Lines 631-632. We have also modified the manuscript to **"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."** Please also see Lines 370-374.

2. Substantial efforts, modification of radiation schemes to interact with cloud droplets (section 2-3), has been put in this paper to study aerosol-cloud interactions during fog simulations. Why did the authors select the fog event that occurred under calm and stable meteorological condition, which is responsible for similar model results between 'S_ARon_CRmod' and 'S_ARon_CRorig' (see last paragraph in section 4-2)? How will simulation results be affected by the modified calculation method of cloud optical property if we choose different fog cases?

Reply: We have described why a fog case was chosen in response to the other reviewer's comments. In the past decades numerical studies on Tule fog have been rare. This is in part due to model's difficulty to simulate fog reasonably. "**We chose this challenging weather system for our first study of this kind since Tule fog is important in safety, hydrology and agriculture in California. 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 which usually do not occur in fog."** The fog case that we chose is a very typical case and thus should be quite representative for a Tule fog event. 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, as the microphysical processes that are involved in fog events are minimal, i.e., only activation and diffusion growth. We are conducting more numerical studies for different type of weather systems using the SOWC model. In this revised version, we have added additional explanation why we chose a fog event for the study. The above bolded text is added in the revised manuscript at Lines 274-278 to explain why the fog phenomena was chosen for our study.

Note: Another unique feature of the modified radiation scheme in our model is that we also consider aerosol radiative properties in the cloud droplets. Most radiation schemes treat cloud droplets are pure water so cloud optical properties only depend on cloud droplet size (radius). However, in our model the optical properties of soot-activated cloud droplets differ from those of sulfate-activated cloud droplets even if they have the same size. Hence, the simulation results can differ not only because of different fog cases but also because of different CCN chemical components, e.g., when one compares a recent fog event with one twenty years ago.

3. Cloud-droplet number concentration between 'S_ARon_CRmod' and 'S_ARon_CRorig' shows significant differences (the difference is greater than the one between 'S_ARon_CRmod' and 'S_ARoff_CRmod'), even though other fields such as Qc, SKT, NSF, LH, and SH are similar between two simulations. Please check the sentences in section 4.2.

Reply: It is true that the difference of the number concentration between 'S_ARon_CRmod' and 'S_ARon_CRorig' is greater than that between 'S_ARon_CRmod' and 'S_ARoff_CRmod'. However, the difference of the cloud mixing ratio amount between the former two experiments (0.001 g m^{-3}) is smaller than that between the latter two experiments (0.007 g m^{-3}). This result stems from the fact that more small cloud droplets were evaporated in S_ARon_CRmod after the use of the new cloud-radiation interaction. We have examined the number concentration of each cloud droplet size between 'S_ARon_CRmod' and 'S_ARon_CRorig' to confirm this finding. Please keep in mind that the difference between 'S_ARon_CRmod' and 'S_ARon_CRorig' is *the calculation of the cloud-radiation interaction* with the same microphysics parameterization, while the difference between 'S_ARon_CRmod' and 'S_ARoff_CRmod' is due to *the neglect of the aerosol-radiation interaction*. The differences of the cloud optical thickness and net downward shortwave radiation between 'S_ARon_CRmod' and 'S_ARon_CRorig' (0.41 for COT and 0.46 for NSF) are also smaller than those between 'S_ARon_CRmod' and 'S_ARoff_CRmod' (1.07 for COT and 3.68 for NSF). Thus, the differences of the meteorological variables (*SKT, NSF, LH, and SH*) between 'S_ARon_CRmod' and 'S_ARon_CRorig' are small.

In the original radiation scheme S_ARon_CRorig, the cloud droplets are assumed to have uniform size; however, in the modified radiation scheme S_ARon_CRmod, cloud droplet size varies for each bin and source types. Additionally, the formula of cloud optical thickness (COT), single scattering albedo and asymmetry factor in the modified radiation scheme are all updated in S_ARon_CRmod. Cloud optical thickness in the original radiation scheme, for example, is a function of cloud water path (CWP) and effective radius (4 μm ≤ r_e ≤ 20 μm) derived from the total droplet number:

$$\tau_{orig}(\lambda) = CWP \times (-6.59 \times 10^{-3} + \frac{1.65}{r_e}). \quad \text{(s1)}$$

However, in the modified radiation scheme cloud optical thickness is a function of cloud droplet size, number, and chemical composition of each bin / source (Eq. 3 in the manuscript). With a similar Q_c, although Q_n 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 would like to emphasize that our main focus of the manuscript is the difference of the aerosol activation between different mixing states (internal vs. source-oriented mixing) and its impact on a fog event, not the modification of cloud-radiation interaction. However, since the size and number concentration of cloud droplets are available from the SOWC model, we elected to use the information to calculate cloud-radiation optical properties even though this level of detail was not readily available when the radiation scheme was first developed.

We agree that the last sentence in section 4.2 could confuse the readers by assuming the difference between S_ARon_CRmod and S_ARon_CRorig only comes from the size distribution. We have modified it to read **"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)."** Please see Lines 487-490. We also add COT in Table 5 in the revised version.

	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**

4. Figure 6 shows that Nitrate concentration in the model is much lower than the observation at all CAAQD stations used in the analysis. How can high Nitrate concentrates in the SJV? What causes high Nitrate concentration in the SJV?

Reply: Nitrate production in the SJV during the winter season primarily occurs via the "dark" chemistry pathway. Background ozone advected into the region from outside California mixes with local NO emissions to form NO_2, NO_3, N_2O_5, and HNO_3 which partitions to condensed particulate nitrate because of cold temperature in the winter. Ying et al. (2009) used a source-oriented air quality model to study source contributions to secondary pollutants formation within California's Central Valley during a severe winter stagnation event during December 2000 – January 2001. In their study, they identified diesel engines as the largest contributor to particle nitrate. Zhang et al. (2014) used the SOWC model to simulate the same episode and studied the effects of particle mixing and feedbacks to meteorology and chemistry without consideration of fogs. More recent simulations for episodes in the year 2010 and later have been unable to reproduce observed nitrate buildup using standalone regional chemical transport models or coupled meteorology-chemical transport models (such as WRF/Chem). The performance of the SOWC model in the current study is typical of such efforts, and considerable new research is directed at improving this feature but results are forthcoming and beyond the scope of the current study.

References:

Ying, Q. and Kleeman, M. J.: Regional contributions to airborne particulate matter in central California during a severe pollution episode, Atmos. Environ., 43, 1218–1228, 2009.

Zhang, H., DeNero, S. P., Joe, D. K., Lee, H.-H., Chen, S.-H., Michalakes, J., and Kleeman, M. J.: Development of a source oriented version of the WRF/Chem model and its application to the California regional PM10 / PM2.5 air quality study, Atmos. Chem. Phys., 14, 485-503, doi:10.5194/acp-14-485-2014, 2014.

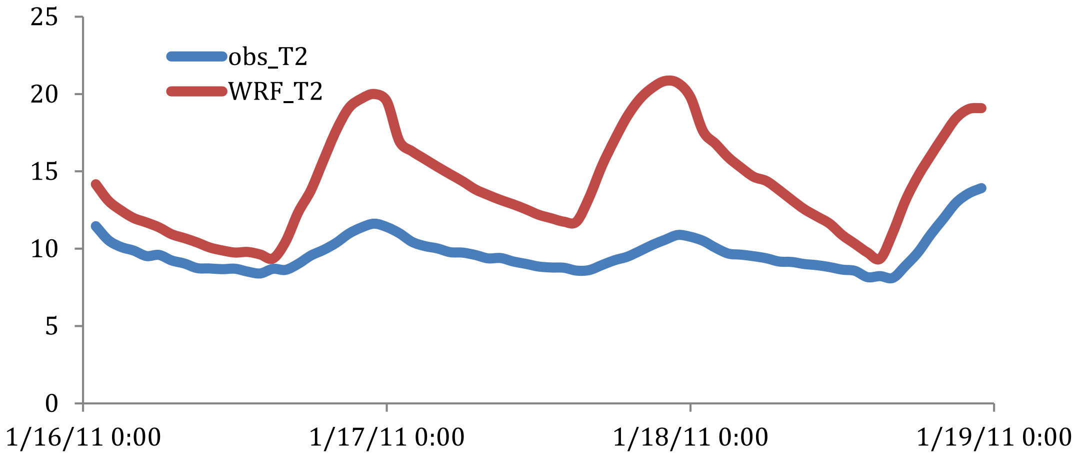
5. Please check specific comments shown below.

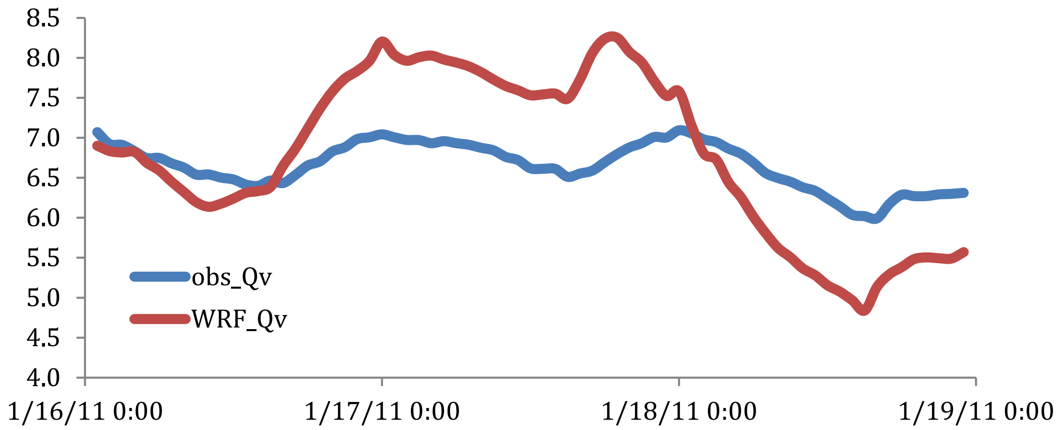
1) It would be better to show available observations for aerosol concentration to compare with the simulated aerosol concentration. Model produces abundant smaller cloud droplets and high CCN concentration, which causes bias in surface temperature.

Reply: Unfortunately, in this study we only have observations available on 18 January, as shown in Figure 6. "**If more discussion of aerosol perditions from the SOWC model is desired, we refer the reader to Zhang et al. (2014) who present a comparison of predicted aerosol concentrations and measured concentrations using field campaign data measured during the California Regional PM_{10} / $PM_{2.5}$ Air Quality Study (CRPAQS) in December 2000 – January 2001**." (Added in the revised manuscript Line 408-412). To address this question using the indirect measurements available in the current study we have conducted two more experiments. The first new experiment uses WRF default prescribed aerosols (chem_opt = 0). Results show that WRF/Chem still underestimated the surface temperature. The surface temperature in daytime was even 1~2 °C colder than that in any experiment in this study due to high aerosol concentration (10^9 #/kg-air; Dp = 10 μm). The second new experiment reduces the prescribed aerosol concentration by one order of magnitude. However, simulated fog was not thick enough in the nighttime and the simulated fog completely disappeared late on 16 January under these test conditions.

The results summarized above confirm that radiative fog indeed is a very challenging weather system for numerical simulations and forecasts. In addition to the potential of too many small cloud droplets, there are two more potential reasons that cause the cold bias in the current study. One is the inaccurate aerosol-cloud-radiation interaction, which is a common and challenging problem in numerical models. The other is the complex terrain in Central Valley of California, where thermodynamical and dynamical processes under fog conditions are difficult to simulate (e.g., drainage flow). Improvements in the aerosol-cloud-radiation interaction in complex terrain is very challenging problem facing all coupled atmosphere-chemistry models.

The figures below provide additional comparison between observations and a WRF simulation, which excludes aerosol direct and indirect effects. The WRF model cannot simulate fog without the inclusion of the aerosol effect. The results show a clear diurnal cycle in simulated 2-m temperature (WRF_T2) because the cloud radiative effect is missing.





References:

Zhang, H., DeNero, S. P., Joe, D. K., Lee, H.-H., Chen, S.-H., Michalakes, J., and Kleeman, M. J.: Development of a source oriented version of the WRF/Chem model and its application to the California regional PM10 / PM2.5 air quality study, Atmos. Chem. Phys., 14, 485-503, doi:10.5194/acp-14-485-2014, 2014.

2) What is the reason of early dissipation of fog in the model simulations?

Reply: The model under-predicts liquid water path in the middle 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 vanishes.**" We have added this explanation in the revised manuscript in Lines 418-419.

3) Which fields are nudged by using FDDA? Temperature and water vapor mixing ratio?

Reply: 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 domain 1 in all simulations.** The bolded texts are added in Lines 376-378.

4) Did you see the same simulation results without KF cumulus scheme in a 4-km inner domain?

Reply: "**No cumulus scheme is used in the most inner domain (4 km resolution).**" It is added in Line 359-360.

5) Please check the following sentence. "aerosol radiative forcing the shortwave energy flux reaching the ground reduces by ~3.7 W m−2 in this case study."

Reply: Modified to **"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"** in Lines 460-461.

6) "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),.." → Even though the authors showed the bias variation (difference) in Figure 9, it would be better to show the diurnal variation of observation and simulation, respectively.

Reply: To limit the number of figures in the manuscript, we added time series variation of T2, Q2 and NSF from observation, S_ARon_CRmod, S_ARoff_CRmod, and I_ARon_CRmod in the supplementary (Figure S1).

7) Please check the following sentences. ", but S_ARon_CRorig had slightly cloud droplet number concentrations (Table 5)."

Reply: Modified to "**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).**" Please also see Lines 487-490.

Supplement

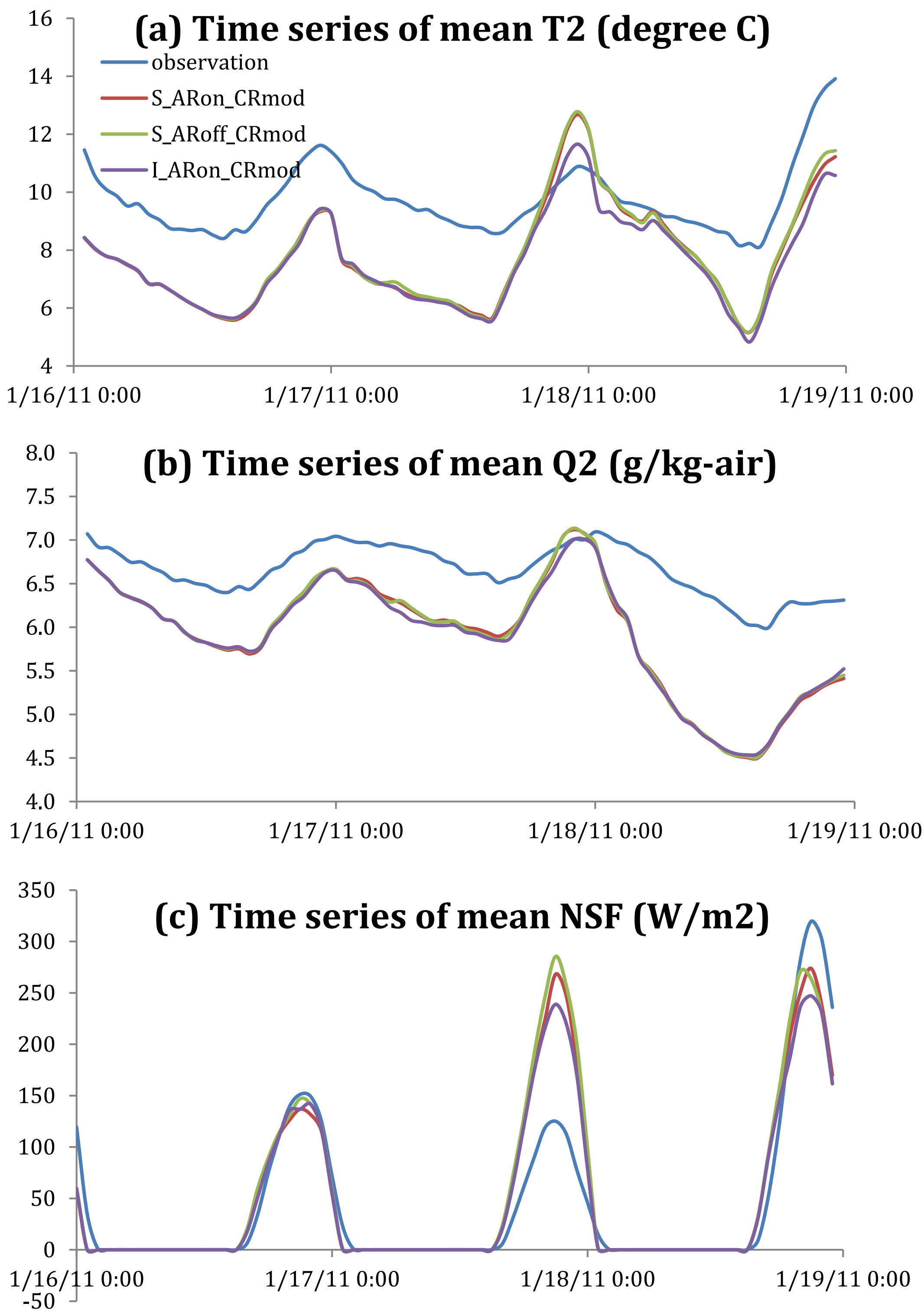


Figure S1. Time series variation of (a) mean 2-m temperature (T2), (b) mean 2-m water vapor mixing ratio (Q2), and (c) mean surface net downward shortwave radiation (NSF) from observations and model simulation from 16 to 18 January 2011.