Review of the manuscript acp-2015-844

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 a s 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.

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 CLAAQC 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.
2. The authors did several model experiments, as defined in Table 3, to investigate how aerosol mixing state influences fog formation. However, they did not outlet clearly what are the specific mechanisms of aerosol influence on fog formation based on the above model results. 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.
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.
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 author 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 author need to synthesize the above-mentioned factors, and give the specific reasons for the simulation results.

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.

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.
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.
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?
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.
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?
6) Line 416-417: What causes are for such an underestimation of LWP? Could this be liked to an underestimation of ammonium nitrate?
7) Line 466-467: How or by what processes does the aerosol direct effect lead to increases of the cloud water mass cloud droplet number?
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;).

Technical issues:

- Line 430-431: Smaller cloud droplets are not shown in the figure.
- Formulas (2)-(5), Tables 4 and 5: Same character Q is used for different variables though having different subscripts.
- Line 463, 574, 589,: Should "water mixing ratio" be a dimensionless variable or it should be called water content?
- Line 544-545: Can one learn hydrophobic nature of particles from Table 6?
- 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?
- Line 617: better to use important "results", instead of "findings".
- Table 1: not fully useful.
- Table 6: What does the Ratio mean? Information on particle size ranges of each bin can be provided.
- Line 1091 (Fig. 7 legend) : should be (b), (d) and (f).
- Line 1123 (Fig. 10): At what supersaturation?
- Line 1154 (Fig. 13): better be plotted in colour.

Referneces

Anttila, T.: Sensitivity of cloud droplet formation to the numerical treatment of the particle mixing stat.: J. Geophys. Res., 115, D21205, doi: 10.1029/2010JD013995. , 2010.

Cubison, M. J., B., Ervens, G., Feingold, K. S., Docherty, I. M., Ulbrich, L., Shields, K., Prather, S., Hering:The influence of chemical composition and mixing state of Los Angeles urban aerosol on CCN number and cloud properties, Atmos. Chem. Phys., 8, 5649–5667, 2008

Kulmala, M., Laaksonen, A., J.Charlson, R., and Korhonen, P.: Clouds without supersaturation, Nature, 388, 336-337, 1997.

Lance, S.,T., Raatikainen, T.B., Onasch, D. R., Worsnop, X.-Y., Yu, M. L., Alexander, M. R., Stolzenburg, P.H., McMurry, J.N., Smith, A., Nenes: Aerosol mixing state, hygroscopic growth and cloud activation efficiency during MIRAGE 2006, Atmos. Chem. Phys., 13, 5049–5062, 2013.

Ma, J. Z., Chen, Y., Wang, W., Yan, P., Liu, H. J., Yang, S. Y., Hu, Z. J., and Lelieveld, J.: Strong air pollution causes widespread haze-clouds over China, J. Geophys. Res., 115, D18204, 10.1029/2009jd013065, 2010.

Makkonen, R., Romakkaniemi, S., Kokkola, H., Stier, P., Raisanen, P., Rast, S., Feichter, J., Kulmala, M., and Laaksonen, A.: Brightening of the global cloud field by nitric acid and the associated radiative forcing, Atmos. Chem. Phys., 12, 7625-7633, 10.5194/acp-12-7625-2012, 2012.

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, 10.5194/acp-14-485-2014, 2014.