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# *Interactive comment on* "Improvement of inorganic aerosol component in $PM_{2.5}$ by constraining aqueous-phase formation of sulfate in cloud with satellite retrievals: WRF-Chem simulations" *by* Tong Sha et al.

**Anonymous Referee #1**

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This paper presents aerosol predictions at Nanjing, China during a haze fog event in which high concentrations of sulfate were observed. The default version of the model underestimated sulfate concentrations and the authors hypothesize that underpredictions in cloud water was a likely explanation. If simulated cloud water is too low it is reasonable to assume the amount of sulfate produced by aqueous chemistry pathways would be reduced. The authors crudely adjust the simulated cloud water to show that sulfate predictions increase when cloud water is increased. This is an obvious result; however, they are correct to note that most air quality studies do not investigate this pathway. Nevertheless, there are flaws in their methodology and analysis that need to be corrected before the paper can be suitable for publication. In addition, there are numerous grammatical errors that make the paper difficult to read. I tried to present suggestions in the abstract, but did not provide corrections for the entire manuscript. The authors should find some assistance to improve the readability of the manuscript.

Specific Comments:

Line 21: Change "it is still a big challenge" to "it is still challenging" or "it is still very challenging".

Line 22: Change "in the numerical model" to "in numerical models"

Line 24: Change "in WRF-Chem" to "in the WRF-Chem model". Change "(November 2018)" to "during November 2018".

Line 25: Change "the sulfate" to "sulfate"

Line 28: Change "dissipation, suggesting that the model" to "dissipation that suggests the model"

Line 30: Change "184 % and 57 %" to "184% and 57%" and similarly elsewhere in the paper.

Lines 30-31: Change "These ultimately result in the simulated SNA 77.2 % higher than the observations." to "These overestimates contribute to the simulated SNA being 77.2% higher than observed."

Line 31-32: Change "However, as the important aqueous-phase reactors, cloud water are simultaneously underestimated by the model." to "However, cloud water is also underestimated by the model which is a pathway for important aqueous-phase reactions."

Line 33: Change "Therefore, the modeled cloud water was constrained" to "Therefore, we constrained the simulated cloud water in a sensitivity simulation"





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Line 36: Change “and can reproduce its diurnal cycles, i.e. the peak concentration at noon” to “and reproduces its diurnal cycle with the peak concentration occurring at noon”

Lines 36-39: This sentence quotes a number which presumably is an average, but then notes differences during nighttime which is awkward. The sentence needs to be reworded.

Line 39: Change “the observation to “the observations”

Line 40: Change “lead” to “leads”. Change “of SNA” to “in SNA”

Line 41: Change “the cloud water” to “cloud water content”

Line 42: Change “bias of SNA simulation” to “simulated SNA bias”.

Line 43: Change “of cloud water can lead to model bias” to “in cloud water content can contribute to model biases”

Lines 43-44: This result is not very surprising but a useful exercise since most air quality simulations tend to focus on uncertainties in emissions, trace gas and aerosol interstitial chemical mechanisms, and boundary layer depth. The effects of clouds are examined less frequently From a process-level perspective, it would be more pleasing to simulate the amount and spatial distribution of cloud water more accurately. That could be achieved in part by constraining the ambient meteorology using data assimilation rather than constraining cloud water content alone.

Lines 113-114: Change “the size distribution” to “droplet size distribution”

Line 133: Change “Model” to “model”.

Line 141: Change “with the horizontal resolution” to “with a grid spacing”. Grid spacing and resolution are different things. A model can resolve phenomena that are bigger than $\sim$5 to 6 times the grid spacing.

Line 142: Change “with 9 km” to “with a grid spacing of 9 km”

Line 144: Change “parameterization schemes” to “parameterizations”. The two words are redundant. The rest of the sentence switches between “parameterization” or “scheme” for unknown reasons. Pick one and be consistent.

Line 151: Change “and Model” to “and the Model”

Line 159: Change “condition” to “conditions”.

Line 162: change “as the” to “used as a”

Lines 171-177: Detail chemistry measurements are only available at two sites, so issues of spatial representativeness need to be address when evaluating the performance of the model. I do not doubt that cloud water is an important factor, but uncertainties in ambient conditions could displace plumes and contribute to model error. The models compares well to surface observations, but there is not assessment of how the model performs aloft which is important as well.

Lines 183-185: State the spatial grid spacing of the observations.

Lines 186-196: While it is encouraging the simulated meteorology is reasonable for one surface site, the authors do not provide the context of what this means in terms of the larger-scale meteorology that is contributing to fog and clouds over a much wider area in which aqueous chemistry is occurring.

Lines 256-257: This is the first place the authors discuss simulated pH. It is not clear in text how they arrived at pH since it is not a standard output of WRF-Chem. In Fig. S2 they state they use an offline ISORROPIA model to compute pH. But that is a different thermodynamic module than the one in MOSAIC, so saying these values are the simulated pH is not correct. So all the assumptions in modeled pH they describe in the manuscript are questionable.

Lines 271-275: The authors describe fog in the text, but the figure is showing LWP which is a vertically integrated quantity. Thus, LWP may not reflect cloud water at the surface. The text implies all the LWP in Figure 5 is fog, but it is possible that parts of those regions are just clouds. If the authors which to describe the areas in Figure 5 as fog, then some additional explanation is needed or different quantities should be plotted.

Lines 276-286: Now the authors focus on surface LWC which is more appropriate in terms of fog. The note that visibility is overestimated by the model, especially on the 27th. But they fail to note that the model seems to underestimate visibility on other days and in other regions, which would argue against their main hypothesis. The amount of sulfate resulting from aqueous chemistry is likely a multiday process so that what is observed at Nanjing is likely an accumulation of sulfate formed over the region that is advected over Nanjing. Another problem with this comparison is that visibility could just be due to high levels of pollution that may be enhanced by aerosol water – and not fog.

Line 298: Differences in the spatial resolution makes it difficult to really assess the potential errors in the model. It would make more sense to average the simulated LWP over the same area as the MODIS grid cells for a more fair comparison. This would affect the fittings performed in the next section.

Line 318: I understand the rationale for modifying the cloud water only in the aqueous chemistry module. But the authors need to put this into context with their findings later in the paper. In reality, changing water content will ultimately impact other aspects of the coupled processes in the model and ultimately change predictions of aerosol species and aqueous chemistry.

Line 397-398: The authors state they investigated the possible reasons contributing to model bias, when in fact they only investigated two: cloud water and specification of pH. The authors need to be more clear and explicit here. The authors could have strengthened their argument by performing other sensitivity simulations that explore other uncertainties such as emissions and chemical mechanisms and putting those results into the context of the cloud water sensitivity analysis.

Lines 431-434: It would have been useful to provide some more reflection of what could be done to improve the cloud predictions. For example data assimilation is often used to constrain the ambient meteorology which often leads to improved cloud predictions. That is a tool that can be used more routinely for air quality applications and is more pleasing than brute force adjusting cloud water content. Exploring other microphysics representation is another option.

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