

Thanks to the referee for his/her very thoughtful suggestions. Below we address the reviewers' comments, with the reviewer comments in black, and our response in blue. We have revised the manuscript accordingly, and mentioned the line number of the **tracked revision**.

Anonymous Referee #3:

General comments:

The chemical transport model is an important tool for the study of air pollution and emission control. The ability of the model to simulate aerosol and its components is an important standard to evaluate the model. This manuscript evaluated the WRF-Chem performance on simulating inorganic aerosol components of PM_{2.5} during a haze-fog event in Nanjing, and investigate the possible reasons of simulating bias compared with the observations. It found that the strong sensitivity of SNA concentration to the cloud water provides an explanation for the bias of SNA simulation. The topic is of interest and the manuscript is generally well written. There are several issues that need to be addressed before the manuscript can be accepted for publication.

Thanks to the reviewer for the comments and suggestions.

Specific Comments:

1. Because LWP is a vertically integrated quantity. Is the large of MODIS LWP possible due to the thickness of the fog is thicker? I think the effect of the vertical profile simulation can be compared. If there is no observation data, vertical sounding and simulation can be compared.

Thanks for suggestions, we agree with the reviewer that the underestimation of LWP may also be caused by the simulated fog thickness being thinner than the observed.

Since we have not found the vertical sounding of cloud water content during this period in Nanjing, so we compared the vertical profile of simulated relative humidity (RH) in the Control run with the observations from Zou et al. (2020) during this haze-fog event (Fig. R1). The observed RH was collected at the Station for Observing Regional Process of the Earth System (SORPES). Due to the lack of other vertical observational data, we set the thickness of fog roughly to a height where the RH is less than 90%. We found that the model underestimates the thickness of fog at 08:00 LT on 26 and 30 November by about 50 m and 150 m but the thickness of simulated fog at 08:00 LT is generally reasonable. Therefore, the deviation of simulated fog thickness is not the main reason that the simulated LWP is lower than the MODIS observations.

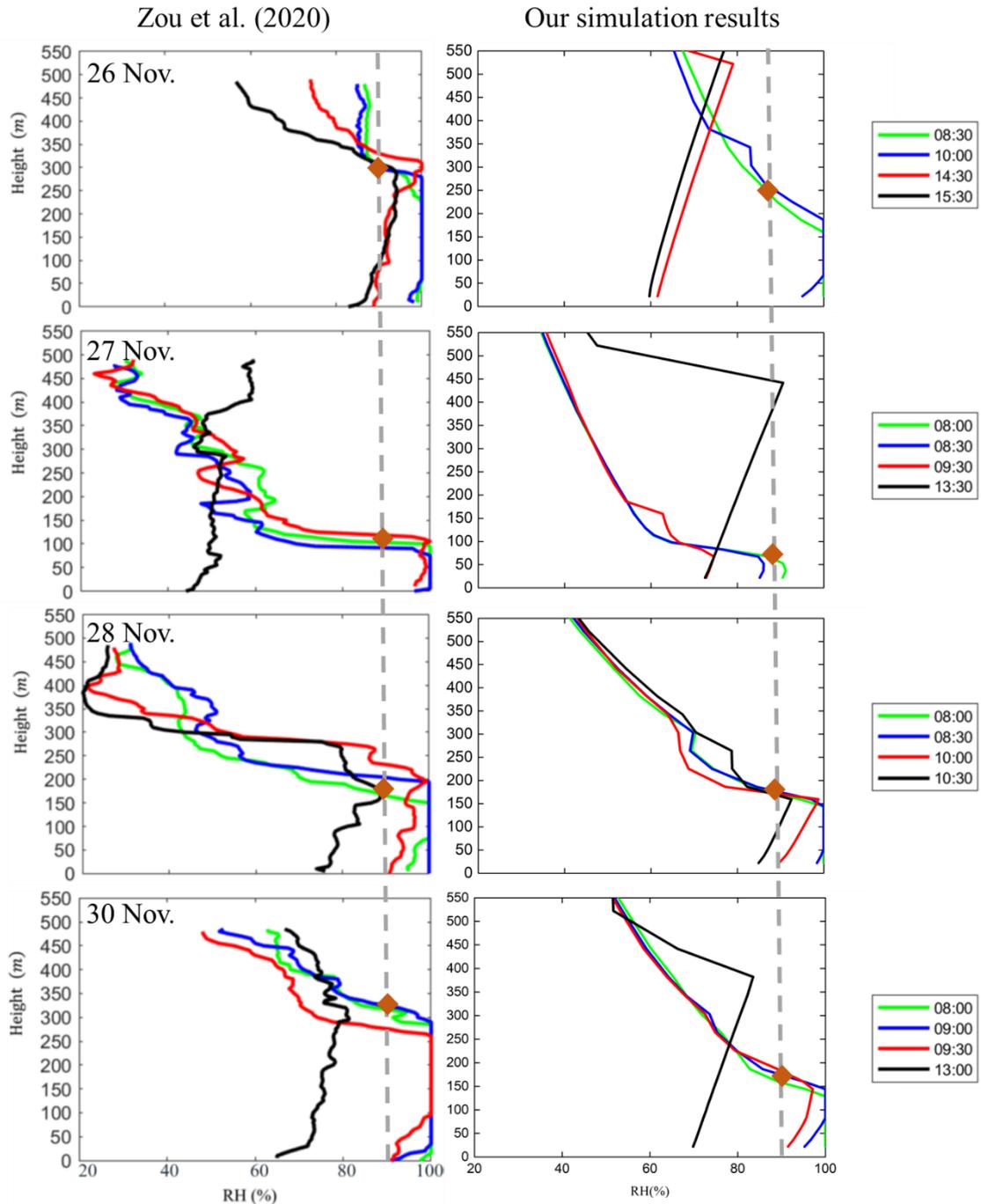


Figure R1. Vertical profile of relative humidity (RH) observed by Zou et al. (2020) and our simulated RH during the four haze-fog days in Nanjing. The different colored lines represent different times. The gray dash lines indicate that the RH is 90%. The brown diamonds present the thickness of fog at 08:00 LT which is the intersection of the vertical profile of RH and RH of 90%.

2. What data quality control did authors do to evaluate the model, especially for Himawari 8 and MODIS?

The AHI/Himawari-8 satellite data has been quality-controlled and validated by the ground testing before releases, so quality control is not required when using the visible

channels of AHI/Himawari-8 satellite data to plot fog areas (Yan et al., 2020; http://www.data.jma.go.jp/mscweb/en/himawari89/space_segment/spsg_ahi.html).

In MODIS Collection 6, the Confidence QA (quality assessment) is set to 3 (i.e., high confidence) for all successful retrievals, so that quality control is no longer required when using this version of the dataset (Platnick et al., 2018). We have added the details in the revised version, see lines 245-249.

Additionally, we compared the spatial distribution of observed LWP from MODIS Collection L3 (MYD08) and L2 (MYD06) cloud products. The resolution of MODIS L2 LWP is $1 \times 1 \text{ km}^2$ (nadir) and was re-gridded to the model grid ($9 \times 9 \text{ km}^2$). Fig. R2 shows that the distribution of LWP from MODIS L2 product is similar to the L3 product during the haze-fog event in the YRD, except on 27 and 30 November, which the location of high LWP in the two products is slightly different. Furthermore, we compare the cumulative probability distribution of LWP from MODIS L3 and L2 products (Fig. R3). The cumulative probability distribution of LWP from MODIS L2 is similar to the L3. The constraining equation for transforming the modelled LWP using the MODIS L2 LWP is:

$$x_m^c = 61.0 \times (x_m + 5.8)^{0.33} + 1.9 \quad (1)$$

Which has a similar constrained function as using the MODIS L3 product (i. e., $x_m^c = 53.0 \times (x_m + 5.8)^{0.4} - 1$). Therefore, LWP from the MODIS L3 product is accurate to evaluate the model and constrain the simulated LWP, and we do not expect an influence of the MODIS resolution on the results herein.

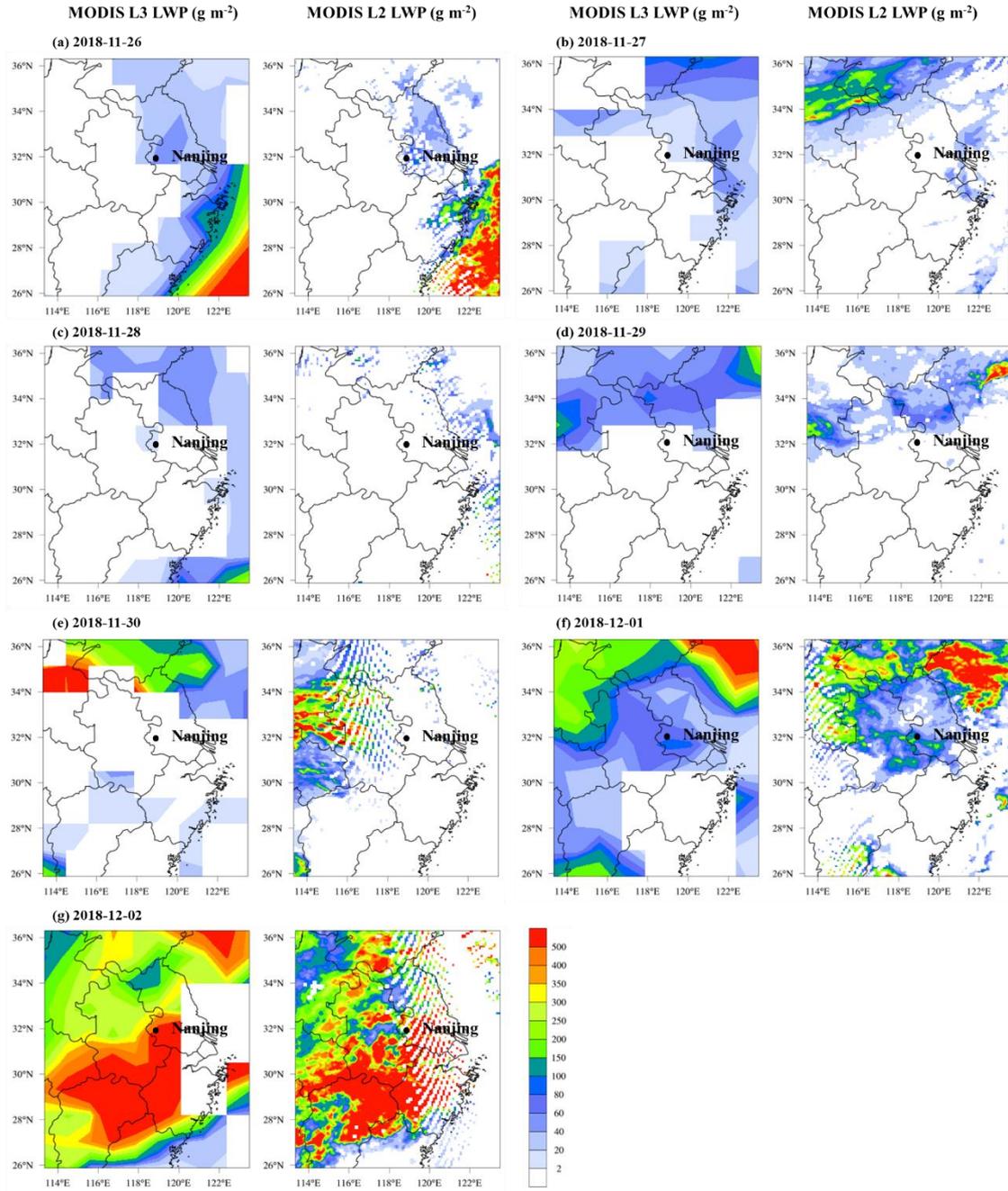


Figure R2. Distribution of the observed LWP (unit: g m^{-2}) from the MODIS Collection 6 Level-3 (columns 1 and 3) and Level-2 cloud product (columns 2 and 4) at 13:30 LT from 26 November to 2 December in the YRD.

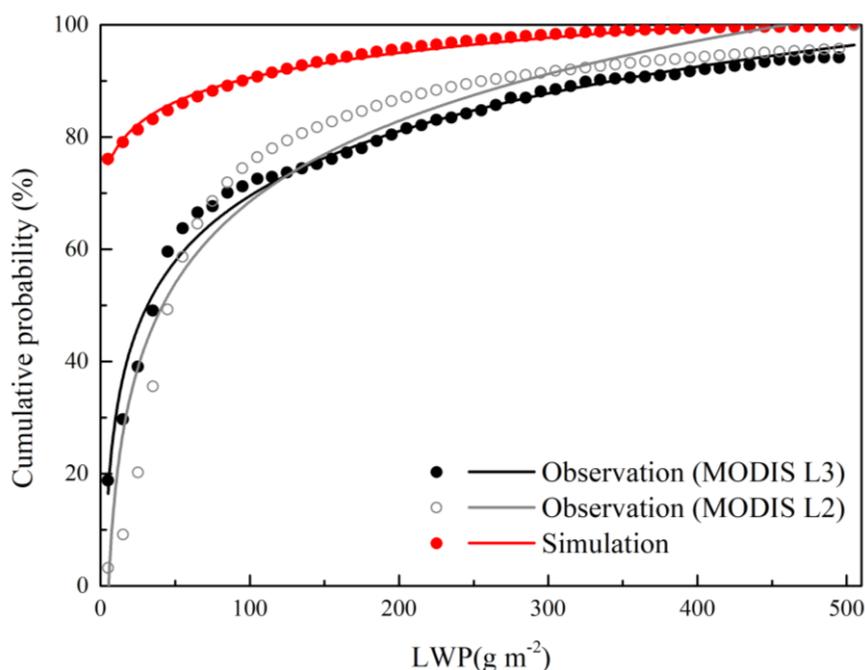


Figure R3. The cumulative probability distribution of LWP from the MODIS observations (L2 and L3 cloud product) and Control simulation (shown as circles). Results are based on statistics of the observed and simulated daily LWP during the haze-fog event in the YRD. The lines present the fitting functions.

3. line 256: What is pH observation data used in this study?

We used the ISORROPIA II (Fountoukis and Nenes, 2007) to calculate the observed $PM_{2.5}$ pH (Fig. S6). The calculation of $PM_{2.5}$ pH is dependent on the concentrations of aerosol components (i.e., Na^+ , SO_4^{2-} , NH_3^+ , NO_3^- , Cl^- , Ca^{2+} , K^+ , Mg^{2+}) and meteorological variables (i.e., RH and temperature).

We have added the details in the revised manuscript, see lines 365-369.

4. Lines 348-350: What the influence of NH_3 and ammonium concentration by changing pH and LWP? Could you provide more detail?

The increase of the cloud water content could enhance the production of sulfate through aqueous-phase chemistry in clouds. More S(VI) concentration can neutralize more NH_3 to form ammonium sulfate ($(NH_4)_2SO_4$) or ammonium bisulfate (NH_4HSO_4), leading to an increase of ammonium concentrations. The hydrogen ion activity in aerosol water can affect the partitioning of TNH_4 between the gas and aerosol phase. Lower aerosol water pH favors TNH_4 toward aerosol ammonium (Weber et al., 2016). See lines 4801-484, 496-500 in the revised version.

5. Lines 365-371: It seems that cloud water pH is important to the aqueous-phase reactions rates, and the model underestimated the cloud water pH in this study. Why the pH was change from 4.9 to 2.5 by modifying LWP? And the authors need to clarify

how to modify the cloud water only in the aqueous chemistry module in this paper. The increase of cloud water content can draw more SO₂ into solution and shift S(IV) speciation towards more reactive forms (H₂SO₃, HSO₃⁻, SO₃²⁻), thus increasing the hydrogen ion concentrations ([H⁺]), and decreasing the cloud water pH (Shen et al., 2012). We have added the above explanations in the revised manuscript, see lines 451-459.

To eliminate the influence of cloud water changes on other processes in the model, we only modify the cloud water content in the aqueous chemistry module, i.e., mosaic_cloudchem_driver.F, and keep the cloud water content in other modules (i.e., microphysics, cumulus parameterization, dry deposition, wet scavenging, photolysis rates, and radiative transfer modules) unchanged. We have added the above descriptions in the revised manuscript, see lines 441-446.

6. In my opinion, emissions, meteorological, and chemistry mechanisms are also main factors in air quality model. The author should make more discussion to prove the importance of they investigated: cloud water and pH. Thanks for suggestions. We agree with the reviewer that meteorological fields, emissions, and chemical mechanisms are also the main factors in air quality models.

Fig. S3 shows the spatial distribution of observed daily SO₂, NO₂, and PM_{2.5} concentrations, as well as the simulated wind speed and direction at 10m from 26 November to 2 December in the YRD. The simulated wind speed at 10m (WS10) is lower than 2 m s⁻¹ in the YRD, and such a low wind speed may not conducive to the advection and diffusion of air pollutants. The easterly wind brings humid air over the ocean. Furthermore, the concentrations of SO₂ and NO₂ mainly produced by primary emissions have no obvious change during this period in Nanjing, indicating that there was no sudden high emission during this event. Therefore, combining the variation of observed air pollutants concentrations and simulated wind fields, the formation of sulfate in Nanjing is mainly attributed to the local chemical reactions, i.e., the in-cloud aqueous-phase chemistry.

We have added more discussions to highlight the importance of in-cloud aqueous-phase chemistry in the revised version, see lines 277-284.

7. Line 815: Describe “WS” twice, change “WS” to “WD”.
Thanks, corrected.

8. Figure 6 should be re-plotted. The circles in the figure could be drawn in larger sizes.
Thanks for suggestions, we have re-plotted it.

Reference:

- Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+ – Ca^{2+} – Mg^{2+} – NH_4^+ – Na^+ – SO_4^{2-} – NO_3^- – Cl^- – H_2O aerosols, *Atmos. Chem. Phys.*, 7, 4639–4659, 2007.
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- Shen, X., Lee, T., Guo, J., Wang, X., Li, P., Xu, P., Wang, Y., Ren, Y., Wang, W., Wang, T., Li, Y., Carn, S. A., and Collett Jr., J. L.: Aqueous phase sulfate production in clouds in eastern China, *Atmos. Environ.*, 62, 502–511, 2012.
- Weber, R. J., Guo, H., Russell, A. G., and Nenes, A.: High aerosol acidity despite declining atmospheric sulfate concentrations over the past 15 years, *Nat. Geosci.*, 9, 282–285, <https://doi.org/10.1038/ngeo2665>, 2016.
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- Zou, J., Sun, J., Li, T., Han, X., Xiang, Z. and Sha, J.: Observed Interactions Among Haze, Fog and Atmospheric Boundary Layer during a Haze-fog Episode in the Yangtze River Delta Region, Eastern China, *Aerosol Air Qual. Res.*, <https://doi.org/10.4209/aaqr.2020.06.0354>, 2020.