Response to Anonymous Referee #2

This study implemented a new aerosol water chemistry module (AWAC) in the WRF-Chem model, and aimed to understand the mechanisms of haze formation over China, in particular, to examine the relative roles of multiphase chemical reactions in aerosol water on particulate sulfate production, which is mainly related to the questions about aerosol pH. They investigated the spatial and temporal distributions of pH around Beijing with the model, and found that the rapid production of sulfate in the NCP can be maintained with the pH range of 4.2-5.7. This is a very interesting and important work. Scientifically, it is still under debate. The analysis of modeling results provided some evidence. However, I still have some questions about the uncertainty of results and the robustness of conclusion. More analysis and clarifications are needed before publication.

We thank the reviewer for the very valuable and constructive comments, which help us to improve the study and manuscript. Please find our point-by-point response (black) and the corresponding revisions (blue and *Italic*) below.

Specific Comments:

1. As the authors also agreed, the pH may be one of the key factors controlling the AWAC processes. However, unfortunately, there is no direct measurement of pH for evaluation. Currently, most studies used the model to calculate the pH, which makes the pH estimation dependent on modules. It is good to couple ISORROPIA II into WRF-Chem, but we still cannot rule out the dependence of pH calculation on this module. In WRF-Chem, the existing module for pH calculation is MOSAIC. Did the authors estimate the pH with MOSAIC and compare the values with ISORROPIA? Are they consistent?

Response:

Good question, thanks. In the original WRF-Chem model Version 3.8, both MADE/SORGAM/ISORROPIA aerosol scheme and MOSAIC aerosol scheme could be used to simulate the aerosol thermodynamics (including aerosol pH and water content). Previous field campaign studies have used the ISORROPIA II model to simulate the aerosol thermodynamics during the haze or non-haze periods in the North China Plain, and reported reasonable model performance (Song et al., 2018;Shi et al., 2017;Liu et al., 2017;Ding et al., 2019;Guo et al., 2017). However, the applicability of MOSAIC aerosol scheme specifically in simulating the aerosol thermodynamics in the North China Plain remains rarely reported. Thus in this study, we have chosen the MADE/SORGAM/ISORROPIA aerosol scheme, and further updated the default ISORROPIA model (Nenes et al., 1998) with the improved version (Fountoukis and Nenes, 2007;Song et al., 2018). The simulated mean pH for different scenarios in our study ranges between 4.2 and 5.7 in the North China Plain, and is comparable with the mean pH values reported in previous relevant studies (e.g., Liu et al., 2017;Shi et al., 2017;Song et al., 2018;Ding et al., 2019).

In the MOSAIC framework (Zaveri et al., 2008), the Multicomponent Equilibrium Solver for Aerosols (MESA) is used to simulate the aerosol thermodynamics (Zaveri et al., 2005a;Zaveri et al., 2005b). The algorithm of MESA model differs significantly from that of ISORROPIA II, including the chemical species involved (Potassium and Magnesium is excluded in MESA), determination of activity coefficient and mutual deliquescence relative humidity (MDRH), and the treatment of phase state and Kelvin Effect. Pye et al. (2020) has compared the pH values estimated by the box-model version of MOSAIC and ISORROPIA II (with the identical modeling input), and they found that the average aerosol pH differed

by 0.3 unit (the difference in pH could be up to 1 unit, and was greater with the decreasing relative humidity). It seems that there is no significant disparity in terms of predicting pH between models of MOSAIC and ISORROPIA. And we agree with the reviewer that it is very important and interesting to compare and analyze the results of different aerosol schemes during the severe haze episodes simulated in our study. But such issue is beyond the research scope of our current study, and it requires considerable efforts to couple the aerosol water chemistry module (AWAC) with the MOSAIC aerosol scheme. Nonetheless, we have added some relevant discussion as a caveat of our study in the Conclusion Section of the revised manuscript:

"Uncertainties relevant with the algorithms to solve the aerosol thermodynamics, including the treatment of non-ideality, size effects, phase state, mixing state, the interactions between inorganic compounds and organic compounds, as well as phase separation, should also be addressed in future studies."

2. For evaluation, since NH3 and NH4+ are so important in this AWAC system, could authors evaluate both of them? In Fig. 1, I didn't find the evaluation of NH4+ and NH3.

Response:

Good suggestion, thanks. Unfortunately, the observational data for ammonia concentrations in the North China Plain during January 2013 is unavailable. Nonetheless we have compared the simulated ammonia concentrations against the observations at other urban Beijing sites during the wintertime in other years:

Table R1. Modelled and observed NH_3, total NH_x (TNH_x) and fraction of NH_x in the particle phase (F_NH4) at urban Beijing sites [a].

	NH_3 mean (ppb)	NH_3 median (ppb)	TNH_x mean (ppb)	TNH_x median (ppb)	F_NH4 mean (%)	F_NH4 median (%)
MEIC_CTRL [b]	4.9	5.0	17.5	12.0	72	70
CTRL [b]	15.5	13.5	28.3	21.2	45	61
Meng et al. (2011) [c]	10.3	/	/		/	/
Liu et al. (2017) [d]	22.0	/	/		/	/
Song et al. (2018) [e]	/	18.0	/	39.1	/	54

[a] The modelling and measuring time differs, including months of November, December, January and February. Nonetheless, estimated emissions and observed concentrations of NH_3 in one study (e.g., Meng et al., 2011;Zhang et al., 2018) both have a minor difference among these months.
[b] Monthly mean value at Tsinghua University site (referred to Beijing site) during January of 2013.
[c] Mean value at Chinese Academy of Meteorological Sciences site during wintertime from 2008 to 2009.
[d] Mean value at Peking University site during November and December in both 2015 and 2016.
[e] Median value at Institute of Atmospheric Physics site from November to December of 2014.

As shown in Table R1, doubling the NH_3 emissions better match the observed ammonia and ammonium concentrations. Furthermore, Kong et al. (2019) estimated that the MEIC inventory (used as the anthropogenic emission inventory in our study) under-predicted NH_3 emissions by about 40% in the North China Plain. Thus, doubling the NH_3 emissions seems a reasonable assumption. We keep Table R1 in the Supplement, and further clarify this issue in the Section 2.2 of the revised manuscript:

"As shown in Table S7 of Supplement, compared with the scenario using the default MEIC emission data, the CTRL scenario (with doubled NH_3 emissions) better matches both the observed ammonia and ammonium concentrations at urban Beijing sites during wintertime (Meng et al., 2011;Liu et al., 2017;Song et al., 2018)."

In Fig. 2, for PM25_OCAT, why not evaluate the absolute values of each components such as K, MG, CA? The emission factor of OCAT is multiplied by 4.5 to match observation. How is this applied? Do you apply it to the total dust emission? This is a huge factor. Did you evaluate the dust mass/AOD over the dust source region to confirm this?

Response:

We thank the reviewer for raising this concern and the very good comment. The dust scheme we used is the improved GOCART dust scheme which is coupled with the MADE/SORGAM aerosol scheme. This dust scheme has been described and evaluated in Zhao et al. (2010) and Zhao et al. (2013). We do not change any of this dust scheme (i.e., the simulated total dust flux is unchanged). WRF/Chem prescribes a specific mass ratio of the fine mode dust emission to the total dust emission, and we further adopt the speciation fractions for the fine mode crustal particles of K, Na, Ca and Mg within the East Asian fine mode dust from Dong et al. (2016). However, Dong et al. (2016) indicated that observed fine particles have a considerably higher mass contribution within the East Asian dust than the chemical transport model (CTM) prescribes. With sensitivity tests, we found that multiply the fine mode dust speciation fractions for K, Na, Ca and Mg by merely one factor of 4.5 could better match the observed PM25_OCAT concentrations at Beijing TSU site.

We have clarified how to tune the PM25_OCAT concentrations in the second and third paragraph of Section 2.2 in the revised manuscript:

"WRF/Chem prescribes a specific mass ratio of the fine mode dust emission to the total dust emission, and we further adopt the fine mode dust emission speciation profiles from Dong et al. (2016). The mass fraction for fine particle components of K, Na, Ca, Mg, Fe and Mn minerals (denoted as PM25_K, PM25_NA, PM25_CA, PM25_MG, PM25_FE and PM25_MN, respectively) from dust source are set as 3.77%, 3.94%, 7.94%, 0.80%, 2.43%, and 0.063%, respectively."

"To match the observations of PM25_OCAT, the emissf_{OCAT} is set to 4.5 (the total dust emission is unchanged), and Dong et al. (2016) indicated that observed fine particles should have a considerably higher mass contribution within the East Asian dust than the chemical transport model prescribes."

Nonetheless, we have compared the simulated AOD_{550nm} with the MODIS as well as AERONET AOD_{550nm} data during January 2013. As shown in Figure R1, the MODIS AOD data is always missing in the vicinity of dust source regions (especially the Gobi Desert), while WRF/Chem simulated an AOD of ~0.2-0.4 there. The model could well reproduce the observed pattern in day-to-day changes of AOD downwind to the Western Pacific. But during the severely polluted episodes, the simulated AOD is not overestimated but rather underestimated especially over the Beijing-Tianjin-Hebei area. Moreover, the simulated AOD at the Dalanzadgad (the capital of SouthGobi Aimag in Mongolia) site is 0.027±0.007, and is also lower than the AERONET AOD data (https://aeronet.gsfc.nasa.gov/cgi-bin/draw_map_display_aod_v3) of 0.076. Results show that multiplying the fine mode dust speciation fractions for K, Na, Ca and Mg by a factor of 4.5 does not lead to the systematic overestimation of AOD. Even though large uncertainties exist for the simulation of dust events, the results of sensitivity tests in our study show that both the diurnal cycle pattern and vertical profile pattern for pH are consistent with varying dust emissions, meanwhile the rapid production of sulfate could be maintained.

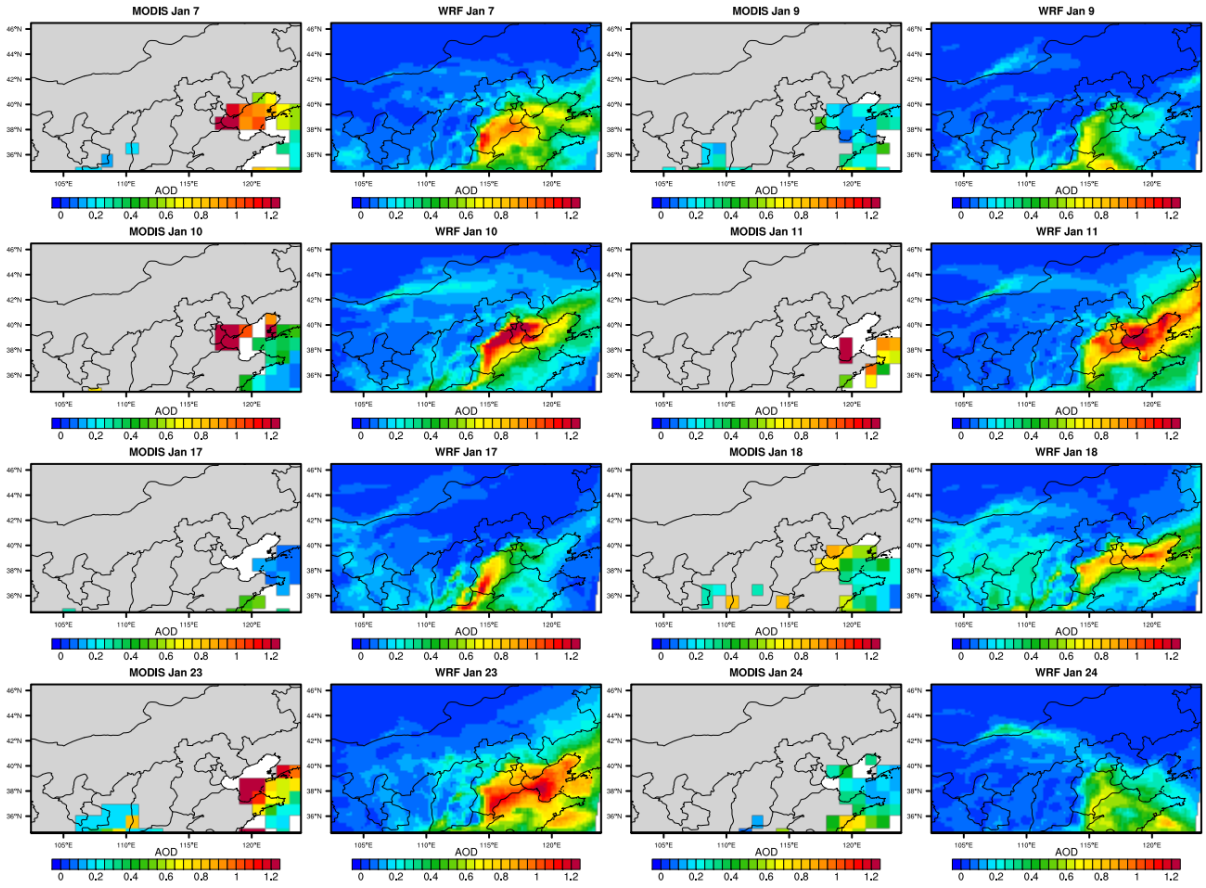

Figure R1. Comparison of MODIS AOD and simulated AOD during January of 2013.

Figure R2 compares the observed PM25_K, PM25_NA, PM25_MG, and PM25_CA with the simulated results (OBS vs. CTRL). The model in general reasonably predicts these individual crustal species, but with a slight overestimation of PM25_CA. We have conducted an additional control simulation tuned to match the observed PM25_K, PM25_MG, PM25_NA and PM25_CA at Beijing TSU site (OCAT_CTRL in Figure R2). The average pH at the surface in OCAT_CTRL case (5.0±0.6) is only about 0.2 unit lower than the CTRL case (5.2±0.5).

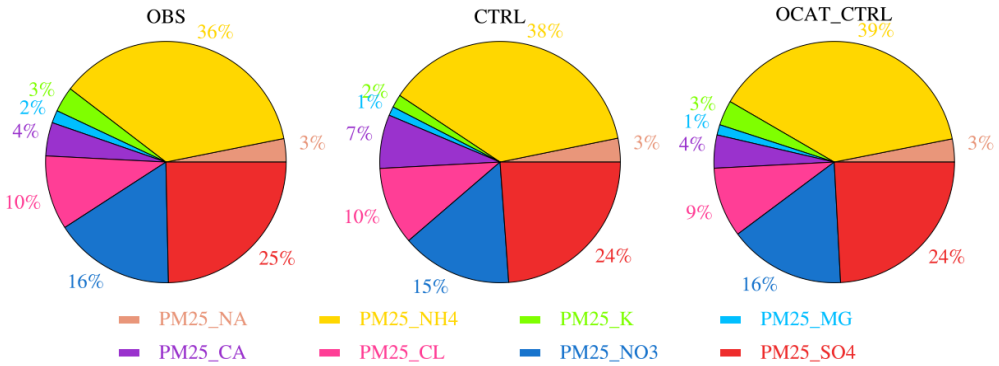

Figure R2. Observed (OBS) and simulated (scenarios of CTRL and OCAT_CTRL) mean electric charge fractions for fine particulate sulfate (PM25_SO4, using SO_4^{2-} as the surrogate), nitrate (PM25_NO3), ammonium (PM25_NH4), chloride (PM25_CL), sodium (PM25_NA), potassium (PM25_K), calcium (PM25_CA) and magnesium (PM25_MG) at TSU site during January of 2013.

3. In Fig. 1 and 2, although the added AWAC significantly increased sulfate production and the mean is closer to the observation, however, it is evident that the model still missed many events. This reflects that there are still some other important processes/mechanisms are missed in the model. Therefore, is it reasonable to use the observation to constrain the model AWAC process? i.e., there may be other processes contributing to the sulfate mass concentration more than AWAC? Please add some explanation and discussion.

Response:

Very good question, thanks. The importance of aerosol water phase production of sulfate has been widely accepted (Li et al., 2017;Chen et al., 2016;Zheng et al., 2015;Cheng et al., 2016;Zhang et al., 2015;Wang et al., 2016;Shao et al., 2019;Xue et al., 2019;Gen et al., 2019;Chen et al., 2019;Wu et al., 2019), and we think that during the severe haze episodes, implementing the heterogeneous reactions in aerosol water might be a key to reduce the gap between modelled sulfate concentrations and observations. After implementing the AWAC, the model performance is significantly improved, and the NMB between observed sulfate-nitrate-ammonium is reduced from -40%~-90% to ±5%. However, as pointed by the reviewer, discrepancies still remain in some events, which may be due to the uncertainties in the treatment of emission, transport (i.e., advection and turbulent mixing), removal (dry and wet deposition), and also the other potential sulfate formation pathways.

Our study focuses on investigating the characterises in the spatio-temporal distribution for aerosol pH as well as sulfate formation budget, and also the uncertainties relevant with assumptions for input parameters. In the CTRL scenario, we tune the input parameters relevant with concentrations for sulfate and other fine particle components (nitrate, ammonium, chlorides and crustal species) to better constrain the spatio-temporal distribution of aerosol pH. The different sources of uncertainties have been tested (Section 3.6). Specifically, we have used the observed sulfate to constrain the TMI formation pathway. Here, we have added one extra simulation of TMI0.5 (both FS_{FE3+} and FS_{MN2+} are halved) to further investigate the uncertainty relevant with TMI concentrations. As shown in Figure 9 of the revised manuscript, our conclusion in the manuscript regarding the diurnal cycle pattern and vertical profile pattern for aerosol pH, as well as the co-existence of multiple sulfate regime and how they interact with pH is consistent in the scenarios of TMI0, TMI0.5 and TMI2.

We have further clarified this issue in the third paragraph of Section 2.2 in the revised manuscript:

"The CTRL scenario is expected to reproduce the observed fine particle compositions (including sulfate, nitrate, ammonium, chloride and crustal components) and gas phase pollutants, and thus more reliably predict the spatio-temporal distribution of pH, AWC and sulfate production … Note that the assumption behind tuning only FS_{FE3+} and FS_{MN2+} to better agree with observed sulfates, is that the model could reasonably simulate the concentrations for other oxidants (e.g., OH, H_2O_2, O_3 and NO_2), thus the deviation from observation can be attributed to the uncertainties in representation of TMI pathway. Note that uncertainties in the emission, transport (i.e., advection and turbulent mixing), removal (dry and wet deposition) and sulfate formation in other phases could also contribute to the discrepancies between modeling results and observations. Nonetheless, this study does not aim at estimating the exact values for aerosol pH and sulfate formation budget. Instead, this study focuses on the characterises in the spatio-temporal distribution for aerosol pH as well as sulfate formation budget, and also the uncertainties relevant with assumptions for input parameters."

Related question, any evidence of significant contribution of AWAC on sulfate production in other events in recent year (2017, 2018, 2019)?

Response:

A very good and interesting question, thanks. The importance of aerosol water phase production of sulfate has been widely discussed and accepted (e.g., Li et al., 2017;Chen et al., 2016;Zheng et al., 2015;Cheng et al., 2016;Wang et al., 2016;Shao et al., 2019;Wu et al., 2019) during the severe haze episodes from early to middle 2010s (Table R2).

Table R2. Selection of the studies focusing on the heterogeneous reactions during the severe haze episodes from early to middle 2010s in China.

Reference	Study time period and area
Zheng et al. (2015)	January 2013 in Beijing–Tianjin–Hebei area
Chen et al. (2016)	October 2014 in North China Plain
Cheng et al. (2016)	January 2013 in Beijing
Wang et al. (2016)	17 November to 12 December of 2012 in Xi'an 21 January to 4 February of 2015 in Beijing
Li et al. (2017)	16 to 27 December 2013 in the Guanzhong basin 13 to 21 January 2014 in Beijing–Tianjin–Hebei area
Shao et al. (2019)	18 October 2014 to 17 January 2015 in the whole China
Wu et al. (2019)	Wintertime of 2015 in the North China Plain

However, the contribution of heterogeneous reactions to sulfate formation in recent years (2017-2020) remains rarely studied and quantified. Note that the air pollution over China has been remarkably mitigated in recent years since the implementation of Clean Air Action in 2013 (Fan et al., 2020;Zhang et al., 2019;Wang et al., 2019;Hou et al., 2019;Cheng et al., 2019). And Zheng et al. (2018) estimated that during 2013–2017, China's anthropogenic emissions decreased by ~60%, ~20% and ~35% for sulfur dioxide (SO_2), nitrogen oxides (NO_x) and $PM_{2.5}$, respectively. Unlike the negative feedback between aerosol loadings and their photochemical production (Kong et al., 2015), the multiphase reactions induce a positive feedback mechanism, i.e., higher particle matter levels lead to more aerosol water, which accelerates sulfate production and further increases the aerosol concentration (Cheng et al., 2016). The role of heterogeneous reactions might exhibit a weakening trend for the inter-annual variation with the decreasing emissions.

4. Line 21 of page 3, "except for" to "besides"?

Response:

Thanks for pointing out this typo, and we have corrected it.

5. Table 2, the description of scenarios includes "halved". It seems to me that there are only two cases: zero and doubled.

Response:

Thanks for pointing out this typo, and we have corrected it.

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