Second Review of "Summertime fine particulate nitrate pollution in the North China Plain: Increasing trends, formation mechanisms, and implications for control policy" by Wen, L., et al.

L. Wen and co-authors have appropriately, thoughtfully, and thoroughly addressed comments and concerns raised in the first review. Below are additional comments on the updated manuscript. Many are editorial in nature, but a few minor details listed below should be addressed prior to acceptance.

Minor Comments:

Page 7: line 12 – 7:15 – Additional model and chemical reaction information provided by the authors during revision is very helpful. Please also provide an estimate of the N_2O_5 uptake coefficient included in the model and briefly compare to other field-studies. Even though $\gamma(N_2O_5)$ is not explicitly included in the mechanism, I believe this uptake coefficient can be estimated from the $N_2O_5(g) \leftrightarrow N_2O_5(a)$ rate constant, using: $k\ (s^{-1}) = 0.25*\gamma(N_2O_5)*SA*c$, where SA is the aerosol surface area and c is the mean molecular speed. I suggest adding this information as it will put the magnitude of N_2O_5 hydrolysis in this study into context of previous studies.

7:16 – What does the model assume for the deposition of nitrate and HNO_3? Dry deposition likely impacts the ground site observations. How do the assumptions pertaining to deposition impact the model results in later sections?

7:29 – What aerosol composition was assumed for the hygroscopic growth calculation?

8:3-8:6 – What method was used for the VOC measurements?

12:19 – How was the early morning period of 06-09:00 LT selected? If the concern is boundary layer expansion and entrainment, this process typically continues past 09:00. If pNO_3^- is mixed down from aloft in the morning (as previous studies have hypothesized), how would this impact the results in this manuscript?

14:7 – What does it mean when the model still predicts nitrate aerosol formation at night when there is no NH_3 present in the model (shown in Figure 7)?

15:2 – Cite Roberts 2008 for the current theory on how particle acidity impacts the yield of $ClNO_2$.

> Roberts, J. M., Osthoff, H. D., Brown, S. S., & Ravishankara, A. R. (2008). N_2O_5 oxidizes chloride to Cl_2 in acidic atmospheric aerosol. *Science, 321*(5892), 1059. https://doi.org/10.1126/science.1158777

16:10 – 16:19 – Thank you to the authors for adding the paragraph on line 17:13. In addition, how sensitive are the results in Figures 7 and 9 to changes in the N_2O_5 gas → particle conversion rate (i.e. uptake coefficient) and $ClNO_2$ formation rate? In theory, if N_2O_5 uptake is inefficient, there will no longer be a linear increase in nitrate with concentrations of O_3 and NO_2 as shown in Figure 7. Have the authors have considered sensitivity tests to these parameters? In addition to

the added paragraph, the authors should also note that the results in Figure 9 only hold if the sensitivity of nitrate production to N_2O_5 uptake does not change under different NOx and O_3 conditions. The authors should also clarify that the model simulations are constrained to ground-based observations and the chemistry aloft may show a different sensitivity than in Figures 7 and 9.

Figure 2 – It might be more helpful to use the "Error bars" to plot the standard deviation of each measurement, not the error in the measurement. That way, the variation in the diurnal average profile can be evaluated. I will leave it up to the authors for what they choose to show.

Editorial Comments:

1:18 – change to "Using historical observations, the nitrate/$PM_{2.5}$ and…"
2:9 – remove "the" before "Earth's"
2:15 – change to "environmental and health consequences, and…"
2:26 – Move "during the day" to after "minimized"
3:4-3:10 – Switch the order of the sentences starting on line 3:4, "Field measurements…" and on line 3:7, "The contribution…".
3:6 – Add McDuffie et al., 2018 and Tham et al., 2018 to the Brown and Stutz reference, since both papers provide overviews of the current state of agreement between field-derived uptake coeffects and laboratory-based parameterizations.

> McDuffie, E. E., Fibiger, D. L., Dubé, W. P., Lopez-Hilfiker, F., Lee, B. H., Thornton, J. A., et al. (2018). Heterogeneous N_2O_5 uptake during winter: Aircraft measurements during the 2015 WINTER campaign and critical evaluation of current parameterizations. *Journal of Geophysical Research: Atmospheres*. https://doi.org/10.1002/2018JD028336
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> Tham, Y. J., Wang, Z., Li, Q., Wang, W., Wang, X., Lu, K., et al. (2018). Heterogeneous N_2O_5 uptake coefficient and production yield of $ClNO_2$ in polluted northern China: Roles of aerosol water content and chemical composition. *Atmospheric Chemistry and Physics Discussions, 2018*, 1-27. https://doi.org/10.5194/acp-2018-313

3:7-3:9 – After the Baasandorj reference, add ", but will be dependent on the rate of NO_3 formation and reaction, and the N_2O_5 uptake coefficient ($\gamma(N_2O_5)$) and formation yield of $ClNO_2$."
3:9 – 3:10 – Add the following references to the Baasandorj reference, which all discuss the vertical transport of nitrate aerosol:

> Brown, S. G., Hyslop, N. P., Roberts, P. T., McCarthy, M. C., & Lurmann, F. W. (2006). Wintertime Vertical Variations in Particulate Matter (PM) and Precursor Concentrations in the San Joaquin Valley during the California Regional Coarse PM/Fine PM Air Quality Study. *Journal of the Air & Waste Management Association*, *56*(9), 1267-1277. https://doi.org/10.1080/10473289.2006.10464583
>
> Prabhakar, G., Parworth, C. L., Zhang, X., Kim, H., Young, D. E., Beyersdorf, A. J., et al. (2017). Observational assessment of the role of nocturnal residual-layer chemistry in

determining daytime surface particulate nitrate concentrations. *Atmospheric Chemistry and Physics, 17*(23), 14747-14770. https://doi.org/10.5194/acp-17-14747-2017

Pusede, S. E., Duffey, K. C., Shusterman, A. A., Saleh, A., Laughner, J. L., Wooldridge, P. J., et al. (2016). On the effectiveness of nitrogen oxide reductions as a control over ammonium nitrate aerosol. *Atmospheric Chemistry and Physics, 16*(4), 2575-2596. https://doi.org/10.5194/acp-16-2575-2016

Watson, J. G., & Chow, J. C. (2002). A wintertime $PM_{2.5}$ episode at the Fresno, CA, supersite. *Atmospheric Environment, 36*(3), 465-475. https://doi.org/https://doi.org/10.1016/S1352-2310(01)00309-0

3:19 – Change to "about a 75% reduction"
4:15 – insert "the" before "North China Plain"
5:15 – Insert "the" before "mountain peak"
5:17 – Change to "descriptions"
6:11 – Change to "quantified *in-situ*"
6:20 – Remove "well qualified and"
6:27 – Remove "the" before "gas-and aqueous…"
7:12 – Add Tham et al., 2018 and McDuffie et al., 2018 from above to the Chang 2011 reference. These studies provide information on the up-to-date status of field-parameterization differences.
7:15 – Change to "utilized previously to simulate…"
8:9 – Clarify what "differences" you are referring to
8:14 – Change to "Simulations were conducted…"
8:16 – Change to "major aerosol formation…"
9:14 – Change to "differences"
9:15 – Change to "differences"
10:1 – Add "power" before "plant"
10:3 – Change "were" to "was"
10:14 – Change to" "thermal decomposition of aerosol"
11:7 – Change to "derived at Mt. Tai from data collected in 2007 and 2014, affirming…" This clarifies that this trend is based on two years of data.
11:18 – Add at the end of the sentence, "at Ji'nan and Mt. Tai, respectively".
11:21 and 11:25 – Subscript NO_x
12:5 – Change "a more and more" to "an increasingly"
13:10 – Change "over" to "out"
16:4 – Remove "to be"
17:7 – Change "that" to "the"
17:17 – Add appropriate references for that statement that increasing aerosol nitrate may reduce the N_2O_5 uptake coefficient.
15:18 – Change "series" to "serious"
Table S1 – Are the units in cm^3 $molecules^{-1}$ s^{-1}? If so, change "mol" to "molec.". If not, disregard this comment.
Figures 8 & 9 – label the three sensitivity regimes on the contour plots