Response to Reviewer 3:

Fine particulate nitrate pollution has been found to play more and more important role in haze pollution in China. This paper reports measurement results of nitrate and relevant species at three distinctly different sites in the North China Plain, the most polluted region in eastern China, and interprets the main daytime and nighttime formation mechanisms of nitrate and discusses its implications for air pollution measures in this region. This paper gives very important insights into the formation mechanisms of summertime fine particulate nitrate and into the control policy of haze pollution in China. It was very well organized and written and can be accepted for publication in ACP as the following points are addressed.

Response: we thank the reviewer for the positive comments and helpful suggestions. We have addressed all of the following points and revised the original manuscript accordingly. For clarity, the referees' comments are listed below in black italics, while our responses and changes in the manuscript are shown in blue and red, respectively.

Major points:

1) The difference between the Mt. Tai and ground surface sites as well as its implication need to be highlighted. The Mt. Tai site locates around 1465 m a.s.l., which is almost near the top of planetary boundary layer (PBL) in summer. This site is not only a "remote site" in this region, but also can provide more insights into the different chemical mechanisms inside or above the PBL, or in the nocturnal PBL and the residual layer. These issue need to be sharpen in the data analysis or in the discussions.

Response: this point raised by the referee is important and constructive. The Mt. Tai data can indeed provide insights into the chemical conditions in the top boundary layer (daytime) and residual layer (nighttime). Our observations at Mt. Tai demonstrate the serious nitrate aerosol pollution throughout the planetary boundary layer in the NCP region. The nitrate formation mechanisms, including the major formation routes and the sensitivities to NOx, O_3 and NH_3, were fairly consistent between Mt. Tai and the surface sites. This suggests the regional homogeneity of the in-situ formation of fine nitrate aerosol within the boundary layer in the NCP region. We have added the following discussion about this issue in the revised manuscript.

"It should be noted that the Mt. Tai site is located at around 1465 m a.s.l., which is almost near the top of PBL in summer. Thus the Mt. Tai data can provide insights into the chemical conditions in the top boundary layer at daytime and in the residual layer during the night. Our observations at Mt. Tai demonstrate the serious nitrate aerosol pollution throughout the PBL in the NCP region. Furthermore, the nitrate formation mechanisms, including the major formation routes and sensitivities to NOx, O_3 and NH_3, were fairly consistent between Mt. Tai and the surface sites. This implies the regional homogeneity in the in-situ formation of fine nitrate aerosol within the PBL over the NCP region."

2) For the MCM modeling of episodes, the model was run at observational-based mode (OBM). Available measurement data, including nitrate, were used as the model inputs. This method of course could help identify the ongoing chemical processes in the air masses, but it

is difficult to trace back to the historical contribution of chemical processes. For example, the observed NH_4NO_3, already existed as initial condition, could be converted into HNO_3 through thermodynamics and further cause an "artificial" mechanism from HNO_3 partitioning. Is that possible to do some sensitivity test by removing or reduction the observed nitrate concentration in the MCM OBM? Otherwise, the authors should mention the weakness or uncertainty of the observational-base modelling when they interpret the modeling results.

Response: we are sorry that the original description of the model setup may be not clear. The RACM-CAPRAM model was only constrained by the hourly measurement data of trace gases and meteorological parameters. The measured aerosol ions data such as nitrate, sulfate and ammonium were only used as initial conditions of the model simulation. The model was initialized with the measured nitrate concentration at the beginning of the episodes, and then simulated the formation of nitrate with constraints of other relevant species. Thus, there should be no artificial mechanism from HNO_3 partitioning with such model setup. We have clarified the detailed model setup by the following statements in the revised manuscript.

"The measured aerosol ions data such as nitrate, sulfate and ammonium were only used as initial conditions of the model simulation. The model was initialized with the measured nitrate concentration at the beginning of the episodes, and then simulated the formation of nitrate with constraints of other relevant species."

Minor points:

1) Please use same scale in Y-axis for the comparison of results from different sites, such as Figure 2, Figure 6 and Figure 7. I understand that the authors would like to highlight some peaks in each panel. However, it is more important to make a comparison between different sites.

Response: these figures have been modified as suggested in the revised manuscript.

2) About the trends of nitrate/$PM_{2.5}$ and nitrate/sulfate in Figure 3, can we also show the trends of nitrate, NO_2 and O_3 concentration if the data are also available?

Response: we don't have measurement data for NO_2 and O_3 before 2010 in Ji'nan. For Mt. Tai, the measured summertime O_3 levels in 2014 (75±21 ppbv) were comparable to those in 2007 (72±19 ppbv), but the NO_2 measurements were not available in 2007. The nitrate data were available at both sites, and we have plotted the trends of nitrate concentrations in the figure below. This figure has been provided in the revised supplementary materials.

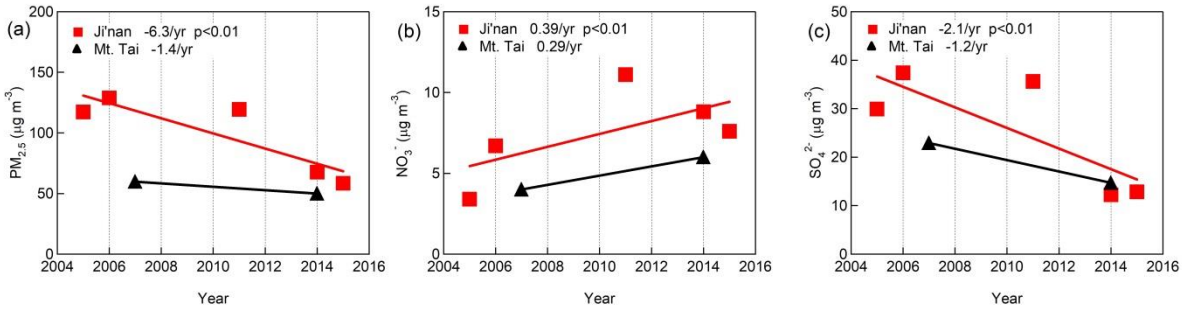

Figure S2. Long-term trends in the absolute concentrations of (a) $PM_{2.5}$, (b) NO_3^-, and (c) SO_4^{2-} in urban Ji'nan and at Mt. Tai in summertime from 2005 to 2015. The fitted lines are derived from the least square linear regression analysis, with the slopes and p values (99% confidence intervals) denoted.

3) References of MARGA measurement: Please add some references of measurements based on this instrument, especially those done in the high aerosol loading environment in China.

Response: two references regarding the deployment of MARGA instrument in the polluted environments of China (Wen et al., 2015; Xie et al., 2015) have been cited in the revised manuscript.

Wen, L., Chen, J., Yang, L., Wang, X., Xu, C., Sui, X., Yao, L., Zhu, Y., Zhang, J., Zhu, T., and Wang, W.: Enhanced formation of fine particulate nitrate at a rural site on the North China Plain in summer: The important roles of ammonia and ozone, Atmos. Environ., 101, 294-302, 2015.

Xie, Y., Ding, A., Nie, W., Mao, H., Qi, X., Huang, X., Xu, Z., Kerminen, V.-M., Petäjä, T., Chi, X., Virkkula, A., Boy, M., Xue, L., Guo, J., Sun, J., Yang, X., Kulmala, M., and Fu, C.: Enhanced sulfate formation by nitrogen dioxide: Implications from in situ observations at the SORPES station, J. Geophys. Res., 120, 12679–12694, 10.1002/2015JD02360, 2015.

4) Page 7, Line 1 and Line 12-14. "Mixing layer height" and "boundary layer height", please use consistent words. In addition, the boundary layer height not only "affects dry deposition", the boundary layer height (or mixing layer height) determines the dispersion capacity of air pollutants emitted from ground surface.

Response: "boundary layer height" has been used in the revised manuscript. We agree that the boundary layer height determines the dispersion capacity of surface air pollutants, but we should note that dispersion was not considered in our box model. The model assumes that the air pollutants are well mixed within the box.

5) Page 9, line 4-5. The uplifted PBL: the developed PBL or uplifted PBL height.

Response: "the developed PBL" was used as suggested.