After reading the comments from you and the reviewers, we have carefully revised our manuscript, and highlighted the changes in yellow. Our responses to the comments are itemized below.

Anything for our paper, please feel free to contact Prof. Gehui Wang via <u>ghwang@geo.ecnu.edu.cn</u>.

All the best

Can Wu On behalf of Prof. Gehui Wang November 11, 2022

Reviewer(s)' Comments to Author:

Reviewer 1

Comments:

I appreciate the authors' revisions to the manuscript and believe it is stronger. In particular, the adding of some meteorological evidence has enhanced the persuasiveness of different physicochemical behaviors between nitrate and ammonium during transport. I think the manuscript is nearly ready for publication and only have minor comments for the authors to further consider.

<u>Reply</u>: We thank the reviewer's valuable comments. We have carefully revised our manuscript according to your advice.

1. <u>Comments:</u>

Please unify the format, "NH₃" or "ammonia".

<u>Reply</u>: Suggestion taken. See page 2, 57-58; page19-20, line 411-437; page 21, line 463; page 24, line 520, 528; page 26, 557;

2. <u>Comments:</u>

Line 270, Please change "Xian" to "Xi'an".

<u>Reply</u>: Suggestion taken. See page 12, line 270.

3. <u>Comments:</u>

Line 290-291, When describing the site, it is suggested to use "Mountain foot" and "Mountainside" uniformly.

<u>Reply</u>: Suggestion taken. We have used the unified expressions for the both sampling sites in the revised manuscript according reviewer's advice. See page 2, line 45-60; page 13, line 290, 294; page 14, line 313; page 23, line 500; page 25, line 538;

4. <u>Comments:</u>

Line 319, What's the differences in $PM_{2.5}$ components at MS site during daylight hours and nocturnal?

<u>Reply</u>: From Figure 7 of the manuscript, we can note that the SNA mainly existed as $(NH_4)_2SO_4$ and NH_4NO_3 both in the daytime and at night, indicating an insignificant difference in the composition of PM_{2.5} that we mainly concerned in this study. However, the sources of daytime and nocturnal PM_{2.5} were different as illustrated the CWT analysis (Figure 4). Above results have been added in the revised manuscript. See page 20, line 431-432.

5. Comments:

*Line 389, What's the relationship between "lower sampling resolution" and "different diurnal cycles between SNA and PM*_{2.5} *at the MF"*?

<u>Reply</u>: As shown in the Figure 3, the hourly PM_{2.5} concentrations exhibited a morning peak at MF site. While, the daily maximum of SNA that collected at 4-hr intervals occurred at 8:00-12:00 LST (Figure S6). This time lag could partially be a result of the lower sampling resolution of SNA that may mask the subtle variation trend in this period.

6. <u>Comments:</u>

Line 487 and Line 521, It's contradictory between "NH₃ emitted from wildfire would be transported aloft and lead to a higher NH₃ and HNO₃ mixing ratio compared to that at lower elevation" and "the source of ammonia sources is unchanged between MS and MF sites".

Reply: The descriptions in line 481-487 are the observation results of the wildfire smoke plumes in the western U.S. (Lindaas et al., 2021), rather than our study. Here we cited this research just for comparing the $P_{HNO3} \times P_{NH3}/Kp$ ratios. During the our campaign, we think that the sources of ammonia was unchanged in the vertical transport process, this can be further verified by organic compounds in the $PM_{2.5}$.As revealed by previous studies (Wang et al., 2006; Wu et al., 2020), the levoglucosan, BkF and IP+BghiP can be used as the tracer for biomass burning, coal combustion and vehicle exhausts, respectively. From Figure S2, the difference in diagnostic ratios and proportion of these organic tracers was indistinctive among two sites. This was indicative of an insignificant change of the corresponding emission sources during the transport. Thus, we think that it is not contradictory between these two descriptions.

7. <u>Comments:</u>

Figure 7: Better to add r^2 *and p values.*

<u>Reply</u>: Suggestion taken. See page 38, line 936.

References

Lindaas, J., Pollack, I. B., Calahorrano, J. J., O'Dell, K., Garofalo, L. A., Pothier, M. A., Farmer, D. K., Kreidenweis, S. M., Campos, T., Flocke, F., Weinheimer, A. J., Montzka, D. D., Tyndall, G. S., Apel, E. C., Hills, A. J., Hornbrook, R. S., Palm, B. B., Peng, Q., Thornton, J. A., Permar, W., Wielgasz, C., Hu, L., Pierce, J. R., Collett, J. L., Jr., Sullivan, A. P., and Fischer, E. V.: Empirical Insights Into the Fate of Ammonia in

Western US Wildfire Smoke Plumes, J. Geophys. Res.-Atmos., 126, 10.1029/2020jd033730, 2021. Wang, G., Kawamura, K., Lee, S., Ho, K., and Cao, J.: Molecular, seasonal, and spatial distributions of organic aerosols from fourteen Chinese cities, Environ. Sci. Technol., 40, 4619-4625, 10.1021/es060291x, 2006.

Wu, C., Wang, G., Li, J., Li, J., Cao, C., Ge, S., Xie, Y., Chen, J., Li, X., Xue, G., Wang, X., Zhao, Z., and Cao, F.: The characteristics of atmospheric brown carbon in Xi'an, inland China: sources, size distributions and optical properties, Atmos. Chem. Phys., 20, 2017-2030, 10.5194/acp-20-2017-2020, 2020.