Dear ACP editor:

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

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

All the best

Can Wu
On behalf of Prof. Gehui Wang
September 30, 2022

Reviewer(s)' Comments to Author:

Reviewer 1

Comments:

The manuscript is a case study on the behaviour of nitrate and ammonium aerosols during transport. Based on the comparison between the chemical composition of MPs sampled simultaneously (with 4h resolution) in a mountain site and a nearby site located at low altitude, the authors describe the processes that soluble inorganic compounds undergo. During transport, the NH_4NO_3 volatilizes, the formed HNO_3 reacts with the dust, forming coarse nitrate, and the available NH_3 reacts with the NH_4HSO_4 forming $(NH_4)_2SO_4$. The isotopic study points to unidirectional reactions and additional ammonia partition into the particulate phases reducing aerosol acidity during transport. A weakness of the article is that some references of interest are missing. A very recent paper on variation of ^{15}N in total particulate N by Wiedenhaus et al., 2021, presents results related to the enrichment of ^{15}N different from those presented in this article, probably because Wiedenhaus et al have *determined ^{15}N for N total. Given the close relationship with the present manuscript, this document should be referenced and similarities/differences should be discussed.*

Reply: We thank the reviewer's valuable comments. We have carefully revised our manuscript according to the comments. The research conducted by Wiedenhaus et al. (2021) has been cited in the revised manuscript, and similarities was also discussed. See page 23, line 492-493, 504-507.

Comments:

The volatilization of NH_4NO_3 and the formation of coarse nitrate is a well-known process by interaction on HNO_3 with coarse sodium and calcium aerosols. Authors did not mention any previous reference to these processes. Authors shall include classical references such as Harrison and Pio 1983, Pakkanen 1996, and others (line 351 and others),

Reply: Suggestion taken. Those references have been cited in the revised manuscript to further support our opinion. See page 17, line 377.

Comments:

The increase in NO_3^- at the mountain site during September 12 and 13 is partially attributed to an external input. This may affect the interpretations of the origin of the nitrate based on the comparison between the averaged concentrations obtained in the studied sites. Therefore, this episode must be excluded for the calculation of the averages used for the interpretation of the modifications of the ions suffered during the transport from the lower levels to the mountain site.

Reply: Suggestion taken. As depicted in the Figure 1, the remaindering data of SNA still exhibited the similar variation trend when excluding the episode occurred during 12-13 September, and the differences in nitrate mass concentration and fractional contribution to $PM_{2.5}$ among two sites become more pronounced. Thus, according to reviewer's advice, the data of this episode were excluded in the subsequent discussion and figures, and we also interpreted this change in the revised manuscript. See page 17, line 365-369.

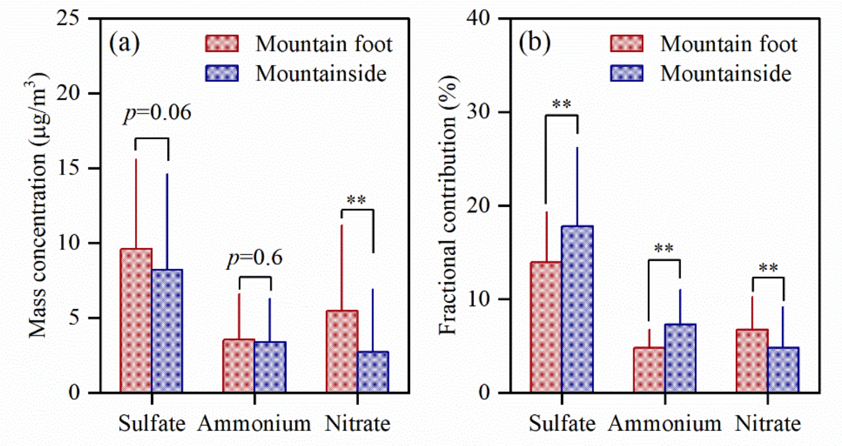

Figure 1 The mass concentration and fractional contributions to $PM_{2.5}$ of SNA excluding the data during 12-13 September at two sites.

Comments:

Ammonia concentrations are not shown. However, some conclusions are based on the differences between the concentrations measured in MF and MS (Lines 407-408). These values, of key interest for the interpretation of data, must be presented or a reference must be included.

Reply: Suggestion taken. The data of NH_3 has been added in the revised manuscript. See pgae 20, line 433-434.

Minor corrections:

Throughout the text, the authors used the term "surface" (surface pollutants…) to refer to the low-elevation site, located in the valley. This is not appropriate. The measurements made at both sites are made at surface level, although at different heights with respect to sea level. I would distinguish between mountain and valley sites, or between high and low elevation sites.

Reply: We agree with the comments above, and modified it in the revised manuscript.

Minor corrections:

Line 92: WHO: add year: WHO, 2021, and add the refence in the reference list:"

Reply: Suggestion taken. See page 30, line 721.

Minor corrections:

Lines 155 and 159: add altitude to the coordinates: e.g. (34º 32'N. 110º5'E, 400 m a.s.l.)

Reply: Suggestion taken. We have added the elevation information of the sampling site as reviewer's advice. See page 7, line 157 and 161.

<u>Minor corrections:</u>

Line 309: delete parenthesis

<u>Reply</u>: Suggestion taken.

<u>Minor corrections:</u>

Line 326: with latitude? Or with altitude?

<u>Reply</u>: Sorry for our carelessness. Here is that the SNA mass fraction increased with altitude, and we have corrected it in the revised manuscript. See page 26, line 344.

<u>Minor corrections:</u>

Figure S2: please, better describe the reason of including Xi'an and Dazhou in Figure S2

<u>Reply</u>: Suggestion taken. As illustrated in Figure 2, the higher CWT loading were mainly distributed in the west and southwest areas of Mt. Hua, indicating that the high SNA aloft was may be mostly driven by the pollution long-range transport from surrounding cities. To further verify above conclusion, we chose these two cities as examples, which situates on the pollution transport pathways and suffered relatively serious haze pollution during 12-13 September. This finding was consistent with our viewpoint, and similar pattern of $PM_{2.5}$ variations also occurred in other cities on the transport pathways, e.g., Xianyang, Weinan (Figure 2). More description has been added in the revised manuscript. See page 17, line 361-365.

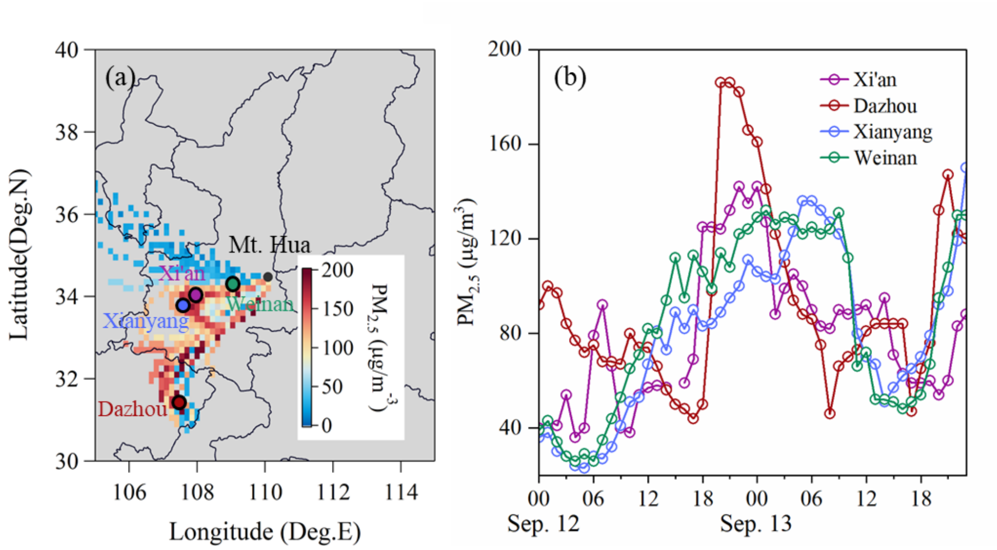

Figure 2 Concentration-weighted trajectory (CWT) analyses of $PM_{2.5}$ during 12-13 Sep. (a). Right panel shows the time series of hourly $PM_{2.5}$ concentrations at different cities on the pollutions transport pathways (b).

Minor corrections:

Line 407: add reference for observational NH_3 data

Reply: Suggestion taken. The NH_3 data observed during the summertime of 2020 has been added in the revised manuscript. See page 20, line 433.

Minor corrections:

Line 453-455: any explanation for these differences?

Reply: The aircraft observations conducted by Lindaas et al. (2021) were mainly focused on the wildfire smoke plumes aloft, which usually contains abundant NH_3 and NO*x*. This would lead to a higher NH_3 and HNO_3 mixing ratio compared to that at lower elevation, and drive a higher $P_{HNO3}×P_{NH3}$/Kp ratio at the upper layers. Above discussions have been added in the revised manuscript. See page 22, line 486-490.

Minor corrections:

Line 460-462 and figure 9: were the samples collected 12-13 September discarded? Origin of pollutant at the two sites may differs during this period.

<u>Reply</u>: Suggestion taken. We have discarded the samples collected during 12-13 September in Figure 7, Figure 9, Figure 10 and Figure S6. As shown in the updated Figure, the recalculated results can also support our viewpoint quiet well.

Reference

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.

Wiedenhaus, H., Ehrnsperger, L., Klemm, O., and Strauss, H.: Stable N-15 isotopes in fine and coarse urban particulate matter, Aerosol Sci. Technol., 55, 859-870, 10.1080/02786826.2021.1905150, 2021.