

## **General.**

We would like to appreciate the editor and reviewers for providing the valuable comments and a better perspective on our work to improve the manuscript. We have taken full account of the comments and revised the manuscript. Responses to specific comments raised by the reviewers are described below. **All the changes made and appeared in the revised text are shown in red. All detailed answers to comments are displayed in blue.**

### **For comments of Referee #2,**

**Response:** We are very grateful for your recognition of this work. In particular, we deeply appreciate the time and effort you've spent in reviewing our manuscript.

### **Comments of Referee #3 and our responses to them**

*This paper summarizes results from observations of aminium species in PM<sub>2.5</sub> aerosols at 11 cities in China, in different regions which experience different meteorology and pollution sources. They identify monomethylammonium, dimethylammonium, and diethylammonium as the dominant species. Their observations demonstrate higher concentrations of aminium species under higher PM<sub>2.5</sub> levels and typically at higher concentrations of acidic aerosol components, suggesting gas to particle partitioning and subsequent acid-base reactions as the main mechanism for aerosol aminium. In some cities, relative humidity and ammonium concentrations appear to control the extent of uptake. The observations are valuable and the content fit a Measurement Report. The authors need to address the following comments before the paper is accepted for*

*publication.*

Response: We appreciate your professional review for our article. We have revised the manuscript to address the comments. Our responses to the specific comments and changes made in the manuscript are given below.

Specific comments:

1) *line 199-200: The higher the Henry's law constant, the more soluble the gas is and higher its partitioning to the aqueous phase. Are the indicated values correct? If so, DEA is the most soluble aminium.*

Response: We are very sorry for the confusion caused by our expression. Indeed, DEA is the most soluble amine. We have updated the relevant sentences.

Lines 201–206: ...the potential of MMA, DMA, and EA to be partitioned into aqueous particles was weaker compared to DEA...Thus, these results imply that the increased emissions of MMA and DMA may partially explain the higher abundance of MMAH<sup>+</sup> and DMAH<sup>+</sup> in PM<sub>2.5</sub> in these investigated cities during winter...

2) *There is a large variability in the reported concentrations of aminium and its ratios to ammonium. Can you please include some statistical analysis to examine if the observed differences in the average concentrations on the polluted vs. clean days significant or not?*

Response: We greatly appreciate your comments. The results of the  $t$ -Test at the 0.05 level for comparing the average value differences of aminium concentrations and its ratios to ammonium during clean and polluted days were added to **Table S1–S3**. In addition, we have updated some statements as follows.

Lines 442–443: ...a large decrease in average  $\text{TA}/\text{NH}_4^+$  and  $\text{TA}/(\text{NH}_3 + \text{NH}_4^+)$  ratios from clean to polluted days occurred in XA ( $t$ -Test,  $P < 0.05$ ) (**Figure 6a, Figure S4, and Table S1–S3**), followed by BJ.

3) *I'm curious why the extent of acidity is defined with 2 times the concentration of sulfate. I believe it's common to calculate acidity ( $\text{H}^+$ ) in molar space for which the molar concentrations of  $\text{H}^+$  associated with sulfate includes the factor of 2 due to its charge balance, but I don't think multiplying the mass concentration by 2 makes sense. Also in Fig. S3 why aren't the organic acids included?*

Response: The correlation analysis of sulfates, nitrates, and organic acids with aminiums has been presented in Figure 4.

We did this (Fig. S3) mainly to facilitate comparison with previous observation (Feng et al., 2022) showing acid as  $[(\text{NO}_3^- + 2\text{SO}_4^{2-}) - \text{NH}_4^+]$  ( $\mu\text{g m}^{-3}$ ). In addition, due to the much lower content of organic acids compared to sulfates and nitrates. We did not include organic acids in Fig. S3.

Reference:

Feng, X., Wang, C., Feng, Y., Cai, J., Zhang, Y., Qi, X., Li, Q., Li, J., and Chen, Y.: Outbreaks of Ethyl-Amines during Haze Episodes in North China Plain: A Potential Source of Amines from Ethanol Gasoline Vehicle Emission, *Environmental Science & Technology Letters*, 9, 306-311, 10.1021/acs.estlett.2c00145, 2022.

Lines 360–361: .... the acidic components (e.g.,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , organic acids, and acidity (expressed as  $[(\text{NO}_3^- + 2\text{SO}_4^{2-}) - \text{NH}_4^+]$  (Feng et al., 2022)),

4) *Line 391: atmospheric oxidation would be a loss pattern for aminium precursors so it's strange to mention this loss process as a possibility to explain the larger concentrations of aminium in aerosols.?*

Response: We thank you for this important comment. We have already deleted this sentence.

5) *figure 4 (and other similar figures): need to add a size legend.*

Response: We used Origin 2021 to create these figures, but we are unable to add size legends in Origin 2021. In the caption, we have emphasized that the larger circle indicates that the correlation is more significant (smaller  $P$ -value), whereas the symbol “×” indicates that the  $P$ -value is greater than 0.05.

6) *I have several editorial suggestions listed below:*

- line 30,39: change “excepting” to “except”
- line 116-117: consider deleting either obviously or inevitably
- line 154: change “determination” to “detection”
- line 304: change “revealed” to “discussed”
- Line 330: “can largely be derived”
- Line 445: change “extraction ratio” to “ratio”
- line 494-498: Modify to “Although a recent study has also demonstrated the possibility of individual aminium species being displaced by ammonia in an environment of high ammonia level (Chen et al., 2022a), the uptake of amines on particles to form aminiums and the mechanisms of relevant influencing factors are still not fully understood”.
- line 507: change “in particular” to “In addition”.

Response: We greatly appreciate your careful review. All the issues mentioned in the above comments have been revised in the revised manuscript.

Lines 30, 39: ...except...

Line 117: ~~inevitably~~

Line 154: detection limits

Line 305: discussed

Line 331: ...can be largely derived from...

Line 444: ~~extraction~~

Lines 493–497: ...Although a recent study has also demonstrated the possibility of individual aminium being displaced by ammonia in an environment of high ammonia level (Chen et al., 2022a), the uptake of amines on particles to form aminiums and the impact mechanisms of relevant influencing factors are still not fully understood...

Line 506: In addition

**Once again, we deeply appreciate the time and effort you've spent in reviewing our manuscript.**