

## **Author responses and changes to Manuscript egosphere-2024-74**

### **“Different Formation Pathways of Nitrogen-containing Organic Compounds in Aerosols and Fog Water in Northern China” by Sun et al.**

The authors are grateful to two anonymous reviewers for their helpful comments and suggestions, which are great advantage to the improvement of the manuscript. The manuscript has been revised thoroughly according to the comments from reviewers. Below, we detail responses and resulting edits to all of the reviewers' comments. We first list the comments in normal font, then followed by our responses in blue. To make it clear, the contents in the revised manuscript are presented in quotes and in italics. We believe that the quality of the manuscript has been greatly improved, and we sincerely welcome further comments from editors and reviewers.

#### **Referee #1**

In this study, the authors investigated the formation pathways of nitrogen-containing organic compounds (NOCs) in both pre-fog aerosols and fog water. One interesting finding is that more and half of NOCs in pre-fog aerosols were originating from primary anthropogenic sources (pNOCs). In addition,  $\text{NH}_3$  addition reactions are likely the important reaction pathways for the formation of secondary NOCs. The paper is well written. The finding of this work provide greater insights into the formation mechanisms of NOCs in the atmosphere. I have a few minor comments and suggestions below.

Reply: Thanks for the positive comments. We have revised the manuscript according to the comments. We hope the revised version can address the concerns from the reviewer.

1. Line 77, “The fog event occurred on the morning of December 10 with visibility <100 m and relative humidity > 90%, lasting from ~06:30 to ~11:20 a.m. (Fig. S1). Two fog water samples (QDF1 and QDF2) were collected using a Caltech Active Strand Cloud water Collector, Version 2 (CASCC2).” The authors shall discuss the potential artifacts when collecting and analyzing the fog samples.

Reply: Thanks for the suggestion. During the campaign, we rinsed the CASCC2 with pure water to obtain a blank sample. The blank sample was processed and analyzed using the same method with samples. The total signal intensities of ion peaks in mass spectrum in blank sample only account for 5% (ESI-) and 14% (ESI+) of those of samples ion peaks on average. Finally, ion peaks in samples with intensities less than 100 times relative to those in the blank sample ion peaks were removed from the formula lists.

We have added the following sentence in the revised manuscript: “*A blank sample for fog water was acquired by rinsing the CASCC2 during the sampling campaign.*” The methods of processing the blank samples have also been clarified: “*Only ion peaks with intensities in sample enhanced >100 times higher than in the blank sample were retained for further analysis.*” Please refer to Lines 84 and 119-120.

2. Line 87, “The duration of sampling for each sample was set as ~23.5 hours, i.e., from 8:00 a.m. to ~7:30 a.m. the next day. Filters were refrigerated at -20 °C immediately after sampling.” Like fog samples, the authors shall discuss the potential artifacts when collecting and analyzing the aerosol samples.

Reply: Thanks for the suggestion. A field blank, which accompanied the field samples to the sampling sites, was prepared during the deployment interval, and then was processed following the same procedure used for the samples. We have clarified it in the revised manuscript. Please refer to Line 93.

3. Line 100, “The neutral molecular formula achieved by adding H (in ESI-) or subtracting H or Na (in ESI+).” Are there other possible adducts for the ions detected in

ESI positive mode?

Reply: Thanks for the reviewer's comment highlighting the needs to clarify our approaches. There are other possible adducts, such as K, during the ionization in ESI+. However, the number of other adducts were very limited. Most of the current studies only considered the addition of H and Na in ESI+ (e.g., Sareen et al., 2016; Jiang et al., 2022). The other adducts were therefore not considered when we assigned formulas. We have clarified it in the revised manuscript. Please refer to Lines 114-117.

### References:

Sareen, N., Carlton, A. G., Surratt, J. D., Gold, A., Lee, B., Lopez-Hilfiker, F. D., Mohr, C., Thornton, J. A., Zhang, Z., Lim, Y. B., and Turpin, B. J.: Identifying precursors and aqueous organic aerosol formation pathways during the SOAS campaign, *Atmos. Chem. Phys.*, 16, 14409-14420, <http://doi.org/10.5194/acp-16-14409-2016>, 2016.

Jiang, H., Li, J., Tang, J., Zhao, S., Chen, Y., Tian, C., Zhang, X., Jiang, B., Liao, Y., and Zhang, G.: Factors Influencing the Molecular Compositions and Distributions of Atmospheric Nitrogen-Containing Compounds, *J. Geophys. Res.: Atmos.*, 127, e2021JD036284, <http://doi.org/10.1029/2021jd036284>, 2022.

4. Line 166, "By comparing the molecular composition of primary anthropogenic emissions (Song et al., 2018; Tang et al., 2020; Song et al., 2021), the NOCs molecules in pre-fog aerosols were categorized into BB, CC, VE, and other sources." Can the authors elaborate what are the other sources? Will some primary NOCs do not classify by this comparison and consider as secondary NOCs?

Reply: Thanks for the constructive comments. The samples were collected at a suburban site in northern China in winter. The site is surrounded by residential area and roads. BB, CC, and VE were identified as the major sources of primary organic aerosols (> 80%) in China (Wu et al., 2018) and significantly contribute to the haze event in northern China (Li et al., 2019). The study conducted at the same site also suggested the significant residential combustion activities (Chen et al., 2023). Therefore, BB, CC,

and VE likely serve as major primary sources at the sampling site. In addition, molecules assigned as “other sources” in this study have high O/C and N/C ratios, which is the characteristics of secondary formation. However, we also totally agree with the reviewer that some primary NOCs (e.g., cooking emission) may not be assigned, because the molecular composition of the other sources cannot be available.

We have clarified them in the revised manuscript: “*Certainly, some other primary sources (e.g., cooking emissions) might also contribute to NOCs, but were not explicitly assigned in this study. This omission could introduce bias into the identification of pNOCs, which warrants more research on the molecular composition of these additional sources in the future.*” Please refer to Lines 189-191.

#### **Reference:**

- Wu, X., Vu, T. V., Shi, Z., Harrison, R. M., Liu, D., and Cen, K.: Characterization and source apportionment of carbonaceous PM<sub>2.5</sub> particles in China - A review, *Atmos. Environ.*, 189, 187-212, <http://doi.org/10.1016/j.atmosenv.2018.06.025>, 2018.
- Li, X., Jiang, L., Bai, Y., Yang, Y., Liu, S., Chen, X., Xu, J., Liu, Y., Wang, Y., Guo, X., Wang, Y., and Wang, G.: Wintertime aerosol chemistry in Beijing during haze period: Significant contribution from secondary formation and biomass burning emission, *Atmos. Res.*, 218, 25-33, <http://doi.org/10.1016/j.atmosres.2018.10.010>, 2019.
- Chen, H., Yan, C., Fu, Q., Wang, X., Tang, J., Jiang, B., Sun, H., Luan, T., Yang, Q., Zhao, Q., Li, J., Zhang, G., Zheng, M., Zhou, X., Chen, B., Du, L., Zhou, R., Zhou, T., and Xue, L.: Optical properties and molecular composition of wintertime atmospheric water-soluble organic carbon in different coastal cities of eastern China, *Sci. Total Environ.*, 892, 164702, <http://doi.org/10.1016/j.scitotenv.2023.164702>, 2023.

5. Line 169, “The number of NOCs derived from BB, CC, and VE is 2298, 1557, and 547, accounting for 61.7%, 41.8%, and 14.7% of the total NOCs, respectively. Some

NOCs were assigned to more than one primary source.” What is the relative contribution of different sources to a specific NOC?

Reply: The source apportionment of NOCs in this study was based on the molecular formula matching. If a specific NOC were assigned to more than one primary source, it would be labeled as “mixed source”. Since the FT-ICR MS is not a quantitative technique, the relative contribution of different sources to a specific NOC cannot be evaluated. However, we primarily focused on the transformation from primary NOCs to secondary NOCs and considered all molecules from BB, CC, and VE as precursors. Therefore, the ambiguous primary sources of some specific NOCs have limited influence on the result.

6. Line 187, “The products of 39 reaction pathways collectively account for 83% of saNOCs, demonstrating their representativeness.” Can the authors further elaborate and specify what are these 39 reaction pathways (e.g. types of reactions and reaction mechanisms)? Any oligomerization and fragmentation reactions consider upon the reactions? How efficient these reaction pathways are in pre-fog and fog event?

Reply: Thanks for the reviewer’s question allowing us to provide more explanation to address the complex reaction pathways. Thirty-nine reaction pathways considered in this study can be classed into eight types, including dealkyl group, oxygen addition, reaction of carboxylic acid, reaction of amine, reaction of the nitro/nitroso group, reaction of sulfur, NH<sub>3</sub> addition, and other reactions. These reactions cover oxidation, hydrolysis, fragmentation, and other typical aqueous-phase reactions. They explained more than 80% of the new-formed NOCs. There are also some other reaction pathways, e.g., oligomerization, were not included in this study, because their corresponding atomic variation within the molecules are too complex to describe by “precursor-product pairs”. We have clarified them in the revised manuscript. Please refer to Lines 209-214.

7. Line 232, "Although it is virtually impossible to identify the functional groups in the formula list obtained by FT-ICR MS, carbonyls have been widely detected in fog

and cloud water (Ervens et al., 2013; Van Pinxteren et al., 2005)." What is the abundance of carbonyls in this study?

Reply: We are sorry for this ambiguous expression. The abundance of carbonyls was not detected in this study. Concentrations of carbonyls (e.g., formaldehyde, glyoxal, and methylglyoxal) in cloud/fog water exhibit considerable variability across different sites (ranging from less than 1 to over 100  $\mu\text{mol L}^{-1}$ ) (Ervens et al., 2013), often constituting over 10% of water-soluble organic carbon (WSOC) in cloud/fog water (Raja et al., 2008; Li et al., 2020b). The WSOC concentrations of two fog water samples in this study were determined to be 226 and 210  $\text{mg C L}^{-1}$ , which is much higher than those in previous studies (Herckes et al., 2013). It can be estimated that the concentration of carbonyls may reach approximately 20  $\text{mg C L}^{-1}$  based on a 10% proportion, significantly surpassing the values at other sites.

#### References:

- Ervens, B., Wang, Y., Eagar, J., Leaitch, W. R., Macdonald, A. M., Valsaraj, K. T., and Herckes, P.: Dissolved organic carbon (DOC) and select aldehydes in cloud and fog water: the role of the aqueous phase in impacting trace gas budgets, *Atmos. Chem. Phys.*, 13, 5117-5135, <http://doi.org/10.5194/acp-13-5117-2013>, 2013.
- Raja, S., Raghunathan, R., Kommalapati, R. R., Shen, X., Collett, J. L., and Valsaraj, K. T.: Organic composition of fogwater in the Texas–Louisiana gulf coast corridor, *Atmos. Environ.*, 43, 4214-4222, <http://doi.org/10.1016/j.atmosenv.2009.05.029>, 2009.
- Li, T., Wang, Z., Wang, Y., Wu, C., Liang, Y., Xia, M., Yu, C., Yun, H., Wang, W., Wang, Y., Guo, J., Herrmann, H., and Wang, T.: Chemical characteristics of cloud water and the impacts on aerosol properties at a subtropical mountain site in Hong Kong SAR, *Atmos. Chem. Phys.*, 20, 391–407, <http://doi.org/10.5194/acp-2019-481>, 2020.
- Herckes, P., Valsaraj, K. T., and Collett, J. L.: A review of observations of organic matter in fogs and clouds: Origin, processing and fate, *Atmos. Res.*, 132-133, 434-449, <http://doi.org/10.1016/j.atmosres.2013.06.005>, 2013.

8. Line 239, “The pH value of fog water in this study was detected as 5.1, while those of pre-fog aerosols were evaluated as less than 4.0 by the ISORROPIA-II model (Zhang et al., 2021).” How the aerosol pH were determined in this study?

Reply: Thanks for the question. The aerosol pH in the study were not determined. However, the sampling site and time of Zhang et al.’s study is as same as this study. They collected PM<sub>1</sub> and PM<sub>>1</sub> during the campaign. The pH values of both PM<sub>1</sub> and PM<sub>>1</sub> collected on Dec. 8 and Dec. 9 were evaluated as < 4.0 by the ISORROPIA-II model. Thus, the pH of PM<sub>2.5</sub> in this study can be expected to < 4.0 as well. We have made a statement in the manuscript where we first used the data from Zhang et al: “*The concentrations of water-soluble ions in pre-fog aerosols were not detected in this study. However, Zhang et al. collected fine (< 1 μm) and coarse particulates (> 1 μm) at the same site.*” Please refer to Lines 263-265.

9. Line 245, "A heatmap plotted based on the molecular classes clearly illustrates the variations in the distribution of pNOCs, saNOCs, and sfNOCs" Would there be a possibility that some primary NOCs (pNOC) may have same chemical formula as secondary NOCs (saNOCs and sfNOCs)?

Reply: Yes, we agree with the comment. The discussion in this study is based on the molecular formulas obtained by FT-ICR MS. A molecular formula may contain multiple isomers. Therefore, the formula assigned as pNOCs may also be formed by secondary processes. However, the formation of these molecules would not increase the richness of organic composition. In this study, we primarily focused on the formation pathways of the new molecular formulas, which increase the chemical richness directly, and the formation of formerly existing molecules were not considered. The above statement has been added in the revised manuscript, please refer to Lines 254-257.

10. Line 256, " Moreover, in this study, the average relative humidity during the pre-fog aerosol collection is  $70 \pm 14\%$  (Fig. S1). Such a high RH may be beneficial for the

formation of HOCs in aerosol liquid water, as evidenced by the observation of aqueous-phase formation of oxygenated organic aerosol during the haze in the North China Plain (Feng et al., 2022; Kuang et al., 2020; Xu et al., 2017)." What are the aerosol water content in this study?

Reply: The concentration of water-soluble ions in pre-fog aerosols was not detected in this study. We used the dataset provided by Zhang et al. to establish the aerosol water content by ISORROPIA-II model. The result showed that PM<sub>1</sub> samples collected in Dec. 8 and Dec. 9 have aerosol water contents of 33.9 and 22.3  $\mu\text{g m}^{-3}$ , respectively. We have added the data in the revised manuscript, please refer to Lines 312-313.

11. Could the authors also comment the concentration and effects of NO<sub>x</sub>, amines and NH<sub>3</sub> on the abundance and formation of NOC in two phases in this study?

Reply: Thanks for the question and suggestion. The following paragraph has been added in the revised manuscript:

*“Finally, gas-phase nitrogen-containing species (NO<sub>2</sub>, amines, and NH<sub>3</sub>) may also influence the NOC formation in aerosols and fog water through two pathways: 1) NOCs formed via gas-phase reactions partition into the condensed phase; 2) gas-phase species partition into the condensed phase and subsequently participate in aqueous-phase NOC formation. While nitration can occur in the gas phase, the concentrations of NO<sub>2</sub> during pre-fog aerosol and fog water collection times were similar (51.5 and 53.0 ppb, respectively). Therefore, the variation in NO<sub>2</sub> levels cannot adequately explain the divergent formation pathways observed in the two phases. Furthermore, reactions between carbonyls and reduced nitrogen predominantly occur in the aqueous phase. Gas-phase NH<sub>3</sub> and amines may influence these reactions through the second pathway. Given the proximity of NH<sub>4</sub><sup>+</sup>/NO<sub>3</sub><sup>-</sup> ratios in aerosols and fog water, variations in NH<sub>3</sub> or NH<sub>4</sub><sup>+</sup> concentrations are unlikely to account for differing reactions in the two phases. This discrepancy may be attributed to an excess of NH<sub>3</sub> in the reactions. However, previous studies have demonstrated that atmospheric amines primarily partition into cloud/fog water (Leng et al., 2015). Importantly, the formation of reduced*



*nitrogen-containing compounds is more efficient in the presence of amines (Jimenez et al., 2022). Thus, gas-phase amines also have the potential to impact NOC formation through partitioning into fog water.”*

Please refer to Lines 286-298.

## References:

Leng, C., Kish, J. D., Roberts, J. E., Dwebi, I., Chon, N., and Liu, Y.: Temperature-Dependent Henry's Law Constants of Atmospheric Amines, *J Phys Chem A*, 119, 8884-8891, <http://doi.org/10.1021/acs.jpca.5b05174>, 2015.

Jimenez, N. G., Sharp, K. D., Gramyk, T., Uglund, D. Z., Tran, M.-K., Rojas, A., Rafla, M. A., Stewart, D., Galloway, M. M., Lin, P., Laskin, A., Cazaunau, M., Pangui, E., Doussin, J.-F., and De Haan, D. O.: Radical-Initiated Brown Carbon Formation in Sunlit Carbonyl–Amine–Ammonium Sulfate Mixtures and Aqueous Aerosol Particles, *ACS Earth Space Chem.*, 6, 228-238, <http://doi.org/10.1021/acsearthspacechem.1c00395>, 2022.

## Referee #2

This paper analyzes the molecular composition of NOCs in both pre-fog aerosols and fog waters, and by comparing with those NOCs from primary emissions, and by using the reaction pathways currently available to assign the NOCs from secondary reactions in aerosols or fog waters to different pathways. The paper is of significance to understand the formation of NOCs in atmospheric condensed phases, therefore the control of NOCs pollution. It is overall well written and the findings are trustworthy with solid evidences, the reviewer has the following minor comments:

Reply: We sincerely appreciate the reviewer’s positive comments. We have responded the reviewer’s concerns as following.

1. One concern is the representativeness of your samples, as you only tested two

aerosol samples and two fog water samples, to assign the NOCs to secondary processes. I fully understand the work load to analyze FT-ICR-MS spectra. You compare the NOCs composition with those from primary emissions which are from previous literatures, and the representativeness of the primary sourced samples has the same limitation. A bit more details are needed. For example, there are many types of biomass, and the NOCs composition might be different in aerosols generated from burning of different materials. This should be carefully explained and justified.

Reply: Thanks for the helpful comments. We totally understand the reviewer's concern about the representativeness of our samples. Although the molecular composition and characteristics of two aerosol or fog water samples is very similar, indicating that the samples can to some extent represent the organic molecular composition of aerosols and fog water during the sampling period, we still clarify the limitation about the sample size: *"It is important to note that our findings may be specific to the observed conditions due to the limited sample size. Therefore, conducting more representative research that includes a broader range of samples under different environmental conditions is necessary in the future."* Please refer to Lines 343-344.

In addition, the biomass burning molecules we used for comparison covered both straw (corn straw and rice straw) and wood (pine branches). Combined with coal combustion and vehicle emission, the sources we considered can cover most of the primary sources at the suburban site during the winter time, as we clarified in Lines 174-177 in the revised manuscript. However, we also realized that some other sources, e.g., cooking, were not considered, which may lead to some bias of primary NOCs identification. We have clarified the limitation in the revised manuscript, please refer to Lines 189-191.

2. saNOCs means NOCs formed in aerosol phases, and those in fog water are formed in aqueous-phase, I am wondering how can you be sure that they are formed in particle phase or aqueous-phase, but not in gas-phase and then condensed on aerosols or partition into aqueous phase? And are these 39 reaction pathways from gas-phase reactions or others? You may need be clear regarding the terminology.

Reply: Thanks for the reviewer's question allowing us to provide more explanation to address the complex reaction pathways. All 39 reaction pathways occur in the aqueous phase. However, some of them can also occur in the gas phase. In 39 reaction pathways, we primarily focused on the nitration and NH<sub>3</sub> addition reactions. Nitration can occur in both gas phase and condensed phase. However, the concentration of NO<sub>2</sub> during pre-fog aerosol and fog water collecting time were close (51.5 and 53.0 ppb, respectively). Therefore, the gas-phase reactions cannot explain the different distribution of NOCs in two phases well. Meanwhile, the reactions between reduced nitrogen and carbonyls mainly occur in the aqueous phase, and the contribution from gas-phase reactions can be expected to be minor. We have added the discussion about the impact of gas-phase species/reactions in the revised manuscript, please refer to Lines 286-298.

3. More information are needed about the 39 reaction pathways, how to choose 39? Any other pathways?

Reply: Thirty-nine reaction pathways considered in this study can be classed into eight types, including dealkyl group, oxygen addition, reaction of carboxylic acid, reaction of amine, reaction of the nitro/nitroso group, reaction of sulfur, NH<sub>3</sub> addition, and other reactions. These reactions cover oxidation, hydrolysis, fragmentation, and other typical aqueous-phase reactions. They explained more than 80% of the new-formed NOCs. There are also some other reaction pathways, e.g., oligomerization, were not included in this study, because their corresponding atomic variation within the molecules are too complex to describe by "precursor-product pairs". The detailed information about the 39 reaction pathways have been added in the revised manuscript, please refer to Lines 209-214.

4. You may need to strength the comparison of your results with other studies that identified NOCs and their sources (both primary and secondary), this part seems to be poor in the current version.

Reply: Thanks for the helpful comments. We have strengthened the comparison with other studies:

- 1) *“Mao et al. found that 28% of CHON- and 32% of CHON+ in aerosols collected at a rural site in North China Plain were common with CC emission (Mao et al., 2022), which is slightly lower than that in this study.”*
- 2) *“A previous study also identified the BB-generated NOCs in ambient aerosols by comparing them with laboratory combustion emissions. A similar distribution of these molecules in the VK plot was observed (Wang et al., 2019).”*
- 3) *“Nitration was widely identified in cloud and fog water (Harrison et al., 2005). However, a recent study showed that aqueous-phase reactions in aerosol liquid water were also important pathways for nitroaromatics formation (Jiang et al., 2023).”*
- 4) *“Numerous laboratory studies have investigated the aqueous-phase reactions between carbonyls and reduced nitrogen (Ervens et al., 2011; Jimenez et al., 2022). However, field evidence of such reactions in the atmosphere remains scarce. Liu et al. have suggested that NH<sub>3</sub> addition reactions are critical in the formation of reduced NOCs in cloud water at Mt. Tai (Liu et al., 2023). The analogous finding in fog water from this study underscores the enhancement of NH<sub>3</sub> addition reactions in dilute aqueous phases.”*

Please refer to Lines 184-186, 195-197, 225-226, and 249-253.

## **References:**

- Mao, J., Cheng, Y., Bai, Z., Zhang, W., Zhang, L., Chen, H., Wang, L., Li, L., and Chen, J.: Molecular characterization of nitrogen-containing organic compounds in the winter North China Plain, *Sci. Total Environ.*, 838, 156189, <http://doi.org/10.1016/j.scitotenv.2022.156189>, 2022.
- Wang, Y., Hu, M., Lin, P., Tan, T., Li, M., Xu, N., Zheng, J., Du, Z., Qin, Y., Wu, Y., Lu, S., Song, Y., Wu, Z., Guo, S., Zeng, L., Huang, X., and He, L.: Enhancement in Particulate Organic Nitrogen and Light Absorption of Humic-Like Substances over Tibetan Plateau Due to Long-Range Transported Biomass Burning Emissions, *Environ. Sci. Technol.*, 53, 14222-14232, <http://doi.org/10.1021/acs.est.9b06152>, 2019.

- Harrison, M. A. J., Barra, S., Borghesi, D., Vione, D., Arsene, C., and Iulian Olariu, R.: Nitrated phenols in the atmosphere: a review, *Atmos. Environ.*, 39, 231-248, <http://doi.org/10.1016/j.atmosenv.2004.09.044>, 2005.
- Jiang, H., Cai, J., Feng, X., Chen, Y., Wang, L., Jiang, B., Liao, Y., Li, J., Zhang, G., Mu, Y., and Chen, J.: Aqueous-Phase Reactions of Anthropogenic Emissions Lead to the High Chemodiversity of Atmospheric Nitrogen-Containing Compounds during the Haze Event, *Environ. Sci. Technol.*, <http://doi.org/10.1021/acs.est.3c06648>, 2023.
- Ervens, B., Turpin, B. J., and Weber, R. J.: Secondary organic aerosol formation in cloud droplets and aqueous particles (aqSOA): a review of laboratory, field and model studies, *Atmos. Chem. Phys.*, 11, 11069-11102, <http://doi.org/10.5194/acp-11-11069-2011>, 2011.
- Jimenez, N. G., Sharp, K. D., Gramyk, T., Uglund, D. Z., Tran, M.-K., Rojas, A., Rafla, M. A., Stewart, D., Galloway, M. M., Lin, P., Laskin, A., Cazaunau, M., Pangui, E., Doussin, J.-F., and De Haan, D. O.: Radical-Initiated Brown Carbon Formation in Sunlit Carbonyl–Amine–Ammonium Sulfate Mixtures and Aqueous Aerosol Particles, *ACS Earth Space Chem.*, 6, 228-238, <http://doi.org/10.1021/acsearthspacechem.1c00395>, 2022.
- Liu, Z., Zhu, B., Zhu, C., Ruan, T., Li, J., Chen, H., Li, Q., Wang, X., Wang, L., Mu, Y., Collett, J., George, C., Wang, Y., Wang, X., Su, J., Yu, S., Mellouki, A., Chen, J., and Jiang, G.: Abundant nitrogenous secondary organic aerosol formation accelerated by cloud processing, *iScience*, 26, 108317, <http://doi.org/10.1016/j.isci.2023.108317>, 2023.

5. I suggest you add a short section to discuss the implication of your findings, so as to highlight the scientific importance of your study.

Reply: Thanks for the constructive suggestion. The following section has been added:  
*“This study highlights that the reactions occurring in the aerosols and fog water yield distinct secondary NOCs compositions. Particularly noteworthy is the identification of NH<sub>3</sub> addition and corresponding reduced NOCs formation in fog water. This*

*phenomenon has not been widely recognized since limited studies have utilized ESI+ to capture reduced NOCs in cloud and fog water (Liu et al., 2023). Both oxidized NOCs (e.g., nitrophenols) and reduced NOCs (e.g., imidazole) constitute brown carbon component. However, their maximum absorption wavelengths differ, potentially resulting in distinct climate effects. Considering that aqueous-phase formed secondary NOCs may contribute to secondary organic aerosols, gaining a deeper understanding of their aqueous-phase formation processes will be crucial for better assessing their climate effects.” Please refer to Lines 345-352.*

**References:**

Liu, Z., Zhu, B., Zhu, C., Ruan, T., Li, J., Chen, H., Li, Q., Wang, X., Wang, L., Mu, Y., Collett, J., George, C., Wang, Y., Wang, X., Su, J., Yu, S., Mellouki, A., Chen, J., and Jiang, G.: Abundant nitrogenous secondary organic aerosol formation accelerated by cloud processing, *iScience*, 26, 108317, <http://doi.org/10.1016/j.isci.2023.108317>, 2023.

6. Some information in the supplement can be put into the main manuscript text

Reply: Thanks for the suggestion. The detailed information about the SPE progress has been moved into the main text. Please refer to Lines 97-104.