Author Response to Comments of the Reviewers

We appreciate the detailed and constructive comments and suggestions from the reviewers. The point-to-point responses to the comments are listed as below.

The *Reviewer comments are black italic font* and the Author responses are blue font.

Author Responses to Reviewer #1

In this manicurist, Xie et al measured the nitrooxy-organosulfates (nitrooxy-OS) in the aerosols during Chinese New Year Eve and aimed at discussing the impact of firework on nitrooxy-OS formation. The paper is well written and in general intriguing. It provides unique information on the molecular characterization, classification and precursors of nitrooxy-OS in ambient aerosols during firework events. This information will help us better understand the formation pathways of nitrooxy-OS and importance of nighttime chemistry/aqueous chemistry under ambient conditions. Therefore, I recommend publication of this manuscript as long as the following concerns are properly addressed.

Response: We really appreciate the valuable comments from the reviewer. We have made changes to both the main text and the supplemental information. Detailed responses are shown below.

1. The title emphasizes "impact of firework", but there is no discussion on "firework" in the abstract part at all. It seems that "impact of firework" is not the whole story of this paper. Either of the title or the abstract should be revised accordingly for consistency of the paper.

Response: The title has been revised to more consistent with the manuscript. Parts of the abstract have also been reformulated (on page 1 lines 1-2; on page 1 lines 20-25).

"Increase of Nitrooxy-organosulfates in Firework-related Urban Aerosols during Chinese New Year Eve"

"High-molecular-weight nitrooxy-OSs with relatively low H/C and O/C ratios and high unsaturation are potentially aromatic-like nitrooxy-OSs. They considerably increased during the New Year's Eve that were affected by the firework emissions. We find that large quantities of carboxylic-rich alicyclic molecules possibly formed by nighttime

reactions. The sufficient abundance of aliphatic-like and aromatic-like nitrooxy-OSs in firework-related aerosols demonstrates that the anthropogenic volatile organic compounds are important precursors of urban secondary organic aerosols (SOA). Besides, more than 98% of those nitrooxy-OSs are extremely low-volatile organic compounds …"

2. Nighttime chemistry is one focus of this manuscript. Can the authors provide more information (e.g., meteorological condition, NOx/O3 concentrations, and if possible, VOC and aerosol chemical composition) to support their discussion on nighttime nitrooxy-OS formation during the focused time period?

Response: The concentrations of chemical components in aerosols have been added in Table S1 in the supplemental information, including water-soluble organic nitrogen (WSON), and water-soluble SO_4^{2-} and NO_3^- (on page 4 lines 7-13).

"They were consistent with the concentration trend of water-soluble organic nitrogen, which were significantly higher in the nighttime than that in the daytime, particularly in NYE N (Table S1) … Meanwhile, the heavy emissions of nitrogen oxide during the firework event could elevate the production rate of NO_3 radicals (Ljungström and Hallquist, 1996; Kiendler-Scharr et al., 2016), and previous study showed a good correlation between NO_3 and the total concentration of nitrooxy-OSs at the night (Nguyen et al., 2014)."

3. Although presented in the table and figures, little discussion is made on the comparison between LNY D and LNYN. It is suggested that a few sentences discussion is added to show the unique situation of firework during the NYE N.

Response: The discussion about the molecular composition in LNY D and LNY N has been made in the manuscript (on page 4 lines 4-5; on page 5 lines 7-8).

"…, similar for the comparison between LNY N (1113) and LNY D (1097) samples, which were in agreement with previous studies…"

"…, and 22 and 23 compounds in LNY D and LNY N, respectively."

4. From Figure 3, it can be concluded that all nitrooxy-OS categories are enhanced during NYE

N. So what category is driven by the firework emission, and what is mainly due to the enhancement of nighttime chemistry? The current discussion is not clear enough.

Response: Thanks. Firework emissions had the most impact on the lignin-like nitrooxy-OSs. The increase of the intensity of carbohydrates-like nitrooxy-OSs is mainly due to the enhancement of nighttime chemistry. The presentation has been added to the manuscript (on page 6 lines 31-33)

"All up, all nitrooxy-OS categories were enhanced in NYE N, particularly for the lignin-like nitrooxy-OSs. Moreover, the intensity of carbohydrates-like nitrooxy-OSs increased due to the enhancement of nighttime chemistry."

References:

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Ljungström, E., and Hallquist, M.: Nitrate radical formation rates in scandinavia, Atmos. Environ., 30, 2925-2932, 1996.

Nguyen, Q. T., Christensen, M. K., Cozzi, F., Zare, A., Hansen, A. M. K., Kristensen, K., Tulinius, T. E., Madsen, H. H., Christensen, J. H., Brandt, J., Massling, A., Nojgaard, J. K., and Glasius, M.: Understanding the anthropogenic influence on formation of biogenic secondary organic aerosols in Denmark via analysis of organosulfates and related oxidation products, Atmos. Chem. Phys., 14, 8961-8981, 10.5194/acp-14-8961-2014, 2014.