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. In particular, we are very grateful to the editor and reviewers for giving us the opportunity to make revision. We have revised our manuscript by fully taking the editor's and reviewers' comments into account. Responses to specific comments raised by the editor and 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.

Comments of Referee #1 and our responses to them

Comments:

The paper presents a comprehensive analysis of the impacts of mist cannon use on air quality in urban areas. While the paper could benefit from improved clarity, the chemical specification seems sound, and the analysis is detailed and thorough. The results of the study are impactful, highlighting the negative effects of mist cannon use on air quality and demonstrating the potential for mist cannons to exacerbate secondary organic aerosol formation and $PM_{2.5}$ pollution in the road environment. These findings have significant implications for future research and policy aimed at improving air quality in urban areas, making this a valuable contribution to the field of air pollution research.

Response: We appreciate the reviewer's valuable comments on our work. Our responses to the specific comments and changes made in the manuscript are given below.

Specific comments:

1) Row 39: The statement about the 13% increase in $PM_{2.5}$ refers to the major effect on health hazard. However, the new compounds potentially formed could be more toxic. The composition change may be more important than the increase in $PM_{2.5}$. Toxicological studies could provide additional information.

Response: We are very sorry for this inappropriate expression. In particular, we greatly appreciate your suggestion. However, due to the research on toxicology seems to deviate from the theme of this study, we have revised the relevant expression.

The relevant revision has been made in the revised manuscript (Lines 39−42).

Lines 39−42: …Thus, the application of mist cannon trucks potentially worsens the road atmospheric environment through the increase in $PM_{2.5}$ level and the production of a large number of water-soluble organic compounds in $PM_{2.5}$…

2) Row 42: This paper lacks proper argumentation why these mist cannon trucks are used in urban environment; how much are the benefits in road dust control?

Response: More descriptions on why the mist cannon trucks are used in urban environment have been added in the revised manuscript. However, specific reports or experimental studies about why and how mist cannon trucks can facilitate road dust control are very scarce. Thus, we conducted this research.

The relevant explanations have been added in the revised manuscript (Lines 57−58 and 63−65).

Lines 57−58: …although the relevant argumentation work is rarely systematically studied or reported…

Lines 63−65: …no study has investigated whether and how the water mist sprayed by mist cannon truck affects the road atmospheric environment…

3) Row 56: Related to previous comment, what are the quantitative benefits of mist cannon use in emission control?

Response: We fully understand your consideration and deeply appreciate your comment. However, as mentioned above, although the mist cannon trucks are considered to be more water-saving and efficient than the traditional sprinkling truck (the ground aspersion), the relevant argumentation work is rarely systematically studied or reported. To our knowledge, this is the first investigation about the effect of mist cannon truck on the formation of water-soluble organic compounds and the pollution control of $PM_{2.5}$.

4) Rows 100-: The experimental description of different roads, sampling sites etc. is difficult to follow based on this text. Insertion of Figure S1 to the main text would help to clarify what has been done.

Response: We appreciate the reviewer's kind suggestion. The Figure S1 has been added in the main text as new Figure 1.

5) Row 109: Can you speciate here how large is the biogenic/traffic VOC roles in your dataset?

Response: Due to the fact that the structure of the determined molecular formula cannot be identified by our analytical method, it is difficult to evaluate the relative importance of biogenic and traffic VOCs in this study. However, this will not affect the main conclusions that were drawn in this study.

6) Row 116: I assume oral fluids are something else than water. Is this a valid background information for your analysis?

Response: We totally agree with your point of view. Thus, the relevant description has been deleted in the revised manuscript.

7) Row 158: Using a noun other than "speed" would be more appropriate in this case.

Response: The sentence has been rewritten in the revised manuscript.

Lines 164–165: ... The samples were injected into the ionization source at 250 μL h^{-1} through a syringe pump…

8) Rows 402-404: It is unclear if the dust stays in the ground or if there are road cleaning

practices to prevent resuspension. Additional information on this would be helpful.

Response: We thank for this important comment. Misting cannon trucks usually operate back and forth on specific road sections to prevent the resuspension of dust. Thus, we need to continuously record the variations in $PM_{2.5}$ mass concentrations near road after the mist cannon truck passed by (Figure 8). More descriptions have been added in the revised manuscript (Lines 452–456).

Lines 451–456: …It should be pointed out that misting cannon trucks usually operate back and forth on specific road sections to prevent the resuspension of dust. After the misting cannon truck passed through the monitoring site several times, repeated online $PM_{2.5}$ monitoring (n = 34, within a month) was performed to exclude the impact of dispersion and traffic flow on analysis results…

9) Row 408: It is unclear what is meant by the "organic" phase in this case.

Response: Several studies have suggested that aerosols can exist in a phase-separated form with an organic shell and an inorganic core even at an RH higher than 80% (Li et al., 2021; Ushijima et al., 2021; Yu et al., 2019). Thus, "organic phase" refers to organic shell in aerosols.

Details were shown in the revised manuscript (Lines 441–443).

Lines 441–443: …This is because aerosols can exist in a phase-separated form with an inorganic core and an organic shell (Yu et al., 2019; Li et al., 2021a; Ushijima et al., 2021) …

10) Row 428: A fraction of road dust is likely in $PM_{2.5}$. It would be helpful to know how much new ambient $PM_{2.5}$ is produced by this operation.

Response: Misting cannon trucks operated back and forth on investigated road sections to prevent the resuspension of dust, as mentioned above. After the misting cannon truck passed through the monitoring point several times, the $PM_{2.5}$ online monitoring was performed to avoid (or decrease) the effect of resuspended road dust. Thus, the resuspension of road dust was expected to exert a relatively minor impact on the $PM_{2.5}$ level near road. The concentration of $PM_{2.5}$ showed an increasing trend after the mist cannon truck passed the monitoring point for 15 minutes. Thus, the water droplets sprayed by the mist cannon truck cannot directly cause an increase in $PM_{2.5}$ concentration, suggesting that the increased $PM_{2.5}$ should be secondarily formed after water mist spraying. This consideration was also supported by a significant increase in the concentration and number of water-soluble organic compounds after air spraying (Fig. 2 and Fig. 3). At 25–35 minutes after the mist cannon truck passed the monitoring point, the increase proportion of $PM_{2.5}$ concentration on the roadside gradually reached the maximum (~13%, on average). Thus, these new ambient $PM_{2.5}$ is secondarily produced by mist cannon truck operation.

More explanations have been added in the revised manuscript (Lines 452–464).

Lines 452–464: …It should be pointed out that misting cannon trucks usually operate back and forth on specific road sections to prevent the resuspension of dust. After the misting cannon truck passed through the monitoring site several times, repeated online $PM_{2.5}$ monitoring (n = 34, within a month) was performed to exclude the impact of dispersion and traffic flow on analysis results. Accordingly, the resuspension of road

dust was expected to exert a relatively minor impact on the $PM_{2.5}$ level near road. The concentration of $PM_{2.5}$ showed an increasing trend after the mist cannon truck passed the monitoring point for 15 minutes. Thus, the water droplets sprayed by the mist cannon truck cannot directly cause an increase in $PM_{2.5}$ concentration, suggesting that the increased $PM_{2.5}$ should be secondarily formed after water mist spraying (~15 minutes). This consideration was also supported by a significant increase in the concentration and number of water-soluble organic compounds after air spraying (Fig. 2 and Fig. 3)…

11) Figure 1: The fonts are extremely small and should be enlarged for readability.

Response: The fonts of Figure 1 (now as Figure 2) have been increased as much as possible.

12) Figure 1: Would there be reference data of "no air spray" & "no ground aspersion" to be added as third bars? This would be the reference to compare with in order to understand the effect of an action.

Response: We are very grateful to the reviewers for this excellent suggestion. In this study, we have set up a reference group, as shown in Figures 2d,h,l. When there was no spraying activity, the concentrations of WOSC, ALW, and other components as well as water-soluble organic compound compositions in $PM_{2.5}$ between these two study sites showed a relatively small difference. Thus, although reference data of "no air spray" and "no ground aspersion" to be added as third bars will be very meaningful, the direct

comparison between air spray case and ground aspersion case is sufficient to achieve the purpose of this study.

13) Figure 4: What does it mean that most points outside different categories (ellipses)?

Response: Figure 4 (now as Figure 5 in the revised manuscript) shows OSc values of unique CHO molecules in WSOM in $PM_{2.5}$ collected from different cases. Based on existing classification method, it is not possible to classify all CHO molecules in Figure 5. However, these unique CHO molecules can at least suggest that the water mist from air spraying can indeed enhance the abundance and diversity of CHO compounds in $PM_{2.5}$.

More descriptions have been added in the revised manuscript (Lines 376–379).

Lines 376–379: …Although it is difficult to classify all CHO molecules in Fig. 5, these identified unique CHO molecules can at least suggest that the water mist from air spraying can promote the formation of CHO compounds and increase their molecular diversity…

14) Figure 6: The format of the figure (smiley face, comments) is more suitable for a PowerPoint presentation than a formal research article.

Response: The Figure 6 (now as Figure 7 in the revised manuscript) has been updated. Smiley face and comments have been deleted.

15) Figure 7: It is unclear what the red continuous line represents. Additional information on this would be helpful.

Response: The Figure 7 (now as Figure 8 in the revised manuscript) has been updated. Now, we use the red dashed line to represent the change in the average concentration of $PM_{2.5}$.

At last, we deeply appreciate the time and effort you've spent in reviewing our manuscript.

References

Li, W. J.; Teng, X. M.; Chen, X. Y.; Liu, L.; Xu, L.; Zhang, J.;Wang, Y. Y.; Zhang, Y.; Shi, Z. B. Organic Coating Reduces Hygroscopic Growth of Phase-Separated Aerosol Particles. *Environ. Sci. Technol.* 2021, 55, 16339−16346.

Ushijima, S. B.; Huynh, E.; Davis, R. D.; Tolbert, M. A. Seeded Crystal Growth of Internally Mixed Organic-Inorganic Aerosols: Impact of Organic Phase State. *J. Phys. Chem. A* 2021, 125, 8668−8679.

Yu, H.; Li, W. J.; Zhang, Y. M.; Tunved, P.; Dall'Osto, M.; Shen, X. J.; Sun, J. Y.; Zhang, X. Y.; Zhang, J. C.; Shi, Z. B. Organic coating on sulfate and soot particles during late summer in the Svalbard Archipelago. *Atmos. Chem. Phys.* 2019, 19, 10433−10446.