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 Reviewer #2 and our responses to them

Comments:

This study investigated the changes in aerosols caused by mist cannon trucks. The topic is interesting and practical in China and fits the scope of the ACP journal. However, some concerns need to be addressed before this manuscript can be considered for publication.

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.

Major Comments:

1) The manuscript needs significant improvement in English editing and figures (including captions). A native English speaker should review the manuscript. For example, in Figure 3, the points in the plot are overlapping, making it difficult to distinguish. The "Air spray" and "ground aspersion road" segments were marked as "(A)" and "(B)," which is the same *as the compound classification "(A) unsaturated aliphatic and (B) unsaturated-like." "Common molecules" were marked only in the (b) panel but also used in a, c, and d. Such issues must be fully addressed before the manuscript can be considered for further processing.*

Response: We appreciate the reviewer's kind and valuable comments. In particular, we are very sorry for the confusion caused by our writing.

---For the comment: A native English speaker should review the manuscript.

Answer: The manuscript has been reviewed by a native English speaker (we have expressed our gratitude to the person who helped us edit the language in the acknowledgement section).

---For the comment: In Figure 3, the points in the plot are overlapping, making it difficult to distinguish.

Answer: The overlapped points in Figure 3 (now as Figure 4 in the revised manuscript) indicate common CHO molecules in WSOM in $PM_{2.5}$ collected from different cases. We want to show that the molecular compositions of CHO compounds in $PM_{2.5}$ in the van Krevelen diagram were scattered across wider ranges in the air spray road segment than in the ground aspersion road segment, particularly in the sunny days (24 and 25 March)

More descriptions to support the above results were shown below (Lines 351–355).

Lines 351–355: …Moreover, common CHO molecules accounted for 39% (sunny day) – 63% (cloudy to sunny day) and 90–95% of CHO molecules in $PM_{2.5}$ collected from the air spray and ground aspersion road segments, respectively (Table 1). In contrast, common CHO molecules contributed 81–85% of CHO molecules in $PM_{2.5}$ collected from two road segments without water spray (I vs II)…

---For the comment: The "Air spray" and "ground aspersion road" segments were marked as "(A)" and "(B)," which is the same as the compound classification "(A) unsaturated aliphatic and (B) unsaturated-like." "Common molecules" were marked only in the (b) panel but also used in a, c, and d.

Answer: All confusing marks or symbols have been revised. For example: two road segments without water spray were marked as "I" and "II", respectively; and the classifications of compounds include unsaturated aliphatic-like (UA), highly unsaturated-like (HU), highly aromatic-like (HA), polycyclic aromatic-like (PA), and saturated-like (Sa) molecules.

2) The authors assumed that the background $PM_{2.5}$ concentrations in the two sampling sites are the same. However, this assumption needs to be validated by comparing the two samplers and the meteorology. Since high-volume samplers can easily be influenced by changes in flow rate and street canyon effects can cause large differences between the two sampling sites, the authors should provide more information and discussion on those aspects. The dispersion effects and changes in traffic flow need to be considered when making the statement that the $PM_{2.5}$ increase 20 minutes later is due to the mist trucks.

Response: This is a crucial question. We are very grateful to the reviewers for this excellent comment. We would like to explain this comment from the following two aspects

1) Two research sites are located on the same road without forks. Moreover, 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, the impact of background $PM_{2.5}$ or meteorological factor on $PM_{2.5}$ compositions or levels was expected to be similar between these two study sites.

2) After the misting cannon truck passed through the monitoring point several times, the $PM_{2.5}$ online monitoring (*n* = 34, within a month) was performed to avoid (or decrease) the effect of resuspended road dust (tightly associated with the dispersion effect and traffic flow). Accordingly, the resuspension of road dust was expected to exert a relatively minor impact on the $PM_{2.5}$ level near road. The concentrations 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). Thus, the overall results can indicate that our statement is reliable.

The added descriptions in the revised manuscript were shown below (Lines 225–227 and 452–464).

Lines 225–227: …It suggested that the impact of background $PM_{2.5}$ or meteorological factor on $PM_{2.5}$ composition or level was similar between these two study sites…

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)…

3) The results are based on qualitative information from FT-ICR MS, so statements such as the increase of SOA should be toned down. Also, the size distribution of the droplets between the two types of water spray can cause a large discrepancy between the two samplers with two $PM_{2.5}$ inlets, as the fog droplet caused by the mist cannon trucks can reach a size of >1 um. The authors should add more discussions on the different sizes of droplet particles.

Response: Partitioning gaseous water-soluble organic compounds to particle phase and subsequent reactions in particle phase are the major formation pathways of atmospheric secondary organic aerosols (SOA) (Lv et al., 2022). Thus, SOA has been replaced with water-soluble organic compounds in the revised manuscript.

For the online monitoring of $PM_{2.5}$ mass concentration, the results suggested that 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. This means that the increased $PM_{2.5}$ should be secondarily formed after water 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). Overall, droplet size is not the main factor affecting $PM_{2.5}$ levels in this study. This may be because the strong light and high temperature during the operation of the mist cannon truck can cause rapid evaporation of water droplets.

More explanations have been added in the revised manuscript, as mentioned in comment 2 (Lines 452–464).

4) The authors should provide more evidence for the statement that the enhanced NO_3 formation with high ALW. The heterogeneous reaction is typically more critical during the nighttime (lines 244-245). Additionally, the dependence of the N_2O_5 take-up coefficient and RH was found to be weak after RH>50% (Wang et al., 2020). In this study, RH is usually much higher (~80%).

Response: In polluted regions, the nitrate aerosol is mainly derived from the following two pathways: (1) the gas-phase oxidation of nitrogen dioxide (NO_2) by hydroxyl radical (•OH) to form nitric acid (HNO_3) (Calvert and Stockwell, 1983) and (2) the heterogeneous hydrolysis of dinitrogen pentoxide (N_2O_5) that was produced from the gas-phase reaction of NO_2 with nitrate radical (NO_3•) on aqueous aerosols (Bertram et al., 2009; Wagner et al., 2013; McDuffie et al., 2019). The gas-phase •OH + NO_2 pathway primarily occurs during the daytime and is mainly influenced by the atmospheric oxidation capacity (Chen et al., 2020; Fu et al., 2020). In this study, the field experiments were conducted during daytime (9:00−13:00 LT) with strong sunlight. Moreover, RH is relatively high. Thus, O_3 can be rapidly photolyzed to form •OH under conditions with abundant H_2O and sunlight (Li et al., 2022). In particular, we found that the concentration of nitrate significantly correlated with that of ALW (P < 0.01, R^2 = 0.7). The overall results suggested that the increase in nitrate concentration after air spraying was tightly with increased ALW.

The added descriptions in the revised manuscript were shown below (Lines 250–257).

Lines 250–257: …As we know, the gas-phase oxidation of NO_2 by hydroxyl radical (•OH) to form nitric acid (HNO_3) is an important pathway for the formation of daytime nitrate aerosol (Fu et al., 2020; Chen et al., 2020). Hydroxyl radical can be rapidly produced by O_3 photolysis under conditions with abundant water vapour and sunlight (as in this study) (Li et al., 2022), which is undoubtedly beneficial for the production of HNO_3. Thus, in the region with large NO_x and ammonia emissions (originated from vehicle exhausts (Yang et al., 2022)), the formation of daytime nitrate aerosol could be promoted by enhanced RH (24–43% of increase) caused by air spraying…

5) The O/C ratios (0.03) in Beijing listed are far lower than the current understanding, which is around 0.3-0.7 (Hu et al., 2017). The authors need to explain this large difference.

Response: We apologize for the writing error. The correct O/C ratio should be 0.33 ± 0.11. The revision has been made in the revised manuscript. In addition, a new case (Hu et al., 2017) has been added in the revised manuscript.

Line 284: … 0.33 ± 0.11…

Lines 286–289: … In addition, another study performed in Beijing showed that the average O/C and H/C ratios of organic aerosols were in the range of 0.47–0.53 and 1.52–1.63, respectively (Hu et al., 2017) …

6) The authors should explain why after the air spray, there were more high C number compounds with lower OS compounds, as shown in Fig S4 a, b, and c. The authors should also consider the difference in the k value for SOA and POA in calculations (Zhao et al., 2015).

Response: The study by Zhao et al. (2015) suggested that smaller particle had a higher κ and a higher degree of oxidation. We know the κ value is very important for the hygroscopicity of particles. However, in this study, we want to show that aqueous-phase oligomerization is a potential path of water-soluble organic compound formation (Fig. S4 a, b, and c; now Fig. S3 a, b, and c in the revised manuscript) (Ma et al., 2022). Because increased ALW may promote the oligomerization of CHO compounds, higher C number of compounds can correspond to lower carbon oxidation state (OSc).

Details were shown in the revised manuscript (Lines 382–384).

Lines 382–384: …we also observed increased oligomerization (e.g., methylglyoxal

($C_3H_4O_2$) to form oligomers ($C_{4–7}H_{6–10}O_5$) in particle phase) of CHO compounds after air spraying (Ma et al., 2022) ...

Minor Comments:

1) Line 455: If only 4 data points were used, such a significance test is not solid anymore since it depends on normally distributed data.

Response: We are very sorry for confusion caused by our description. In the main text, we have indicated that linear regression analysis between ALW (n = 8) and WSOC (n = 8) and WSON (n = 8) was conducted based on 4-day data (R^2 = 0.75 – 0.84, P < 0.01).

The number of data points has been added in the revised manuscript (Lines 234 and 497).

Line 234: …Linear regression analysis for all data showed that the mass concentrations of ALW (n = 8) …

Line 497: …we found that the mass concentration of ALW (n = 8) …

2) Line 379 and Line 385: It should be cited as Su et al., 2021, not Sihui et al., 2021.

Response: Thank you very much for your careful review. The revisions have been made in the revised manuscript (Lines 293, 296, 308, 408, and 413).

3) Line 396: What does "abundant medium" mean? It should be rephrased.

Response: We are very sorry for confusion caused by our description. We want to express that ALW is an abundant medium (Nguyen et al., 2016) that can enhance secondary organic aerosol (SOA) formation. In the revised manuscript, we have deleted the phrase and rewritten the sentence.

Lines 425–426: …the increase in ALW caused by air spraying can facilitate the formation of organic nitrates…

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

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