Dear Editor-in-Chief,

We greatly appreciate the editor for thoroughly examining our manuscript and providing very helpful comments to guide our revision. Here we submit a new version (No.: acp-2022-321), entitled: "**Measurement Report: Investigation of pH- and particle size-dependent chemical and optical properties of water-soluble organic carbon: implications for its sources and aging processes**". We have carefully addressed all the comments provided by the editor. In the attachment, an item-by-item response to the comments of the editor is given below. All revisions are highlighted in blue color in the main text of the revised manuscript.

Thank you for taking care of the review process for this paper.

Sincerely,

Prof. Jihua Tan and coauthors
College of Resources and Environment, University of Chinese Academy of Sciences, Beijing 100049
tanjh@ucas.ac.cn

Editor's Comments

Based on the comments of the expert in the field and myself, and after my consideration, the manuscript is of adequate interest to merit publication in Atmospheric Chemistry and Physics as a Measurement report. The authors have thoroughly responded all the questions/comments raised by the reviewer and me, and modified the manuscript according to the suggestions and important changes have been done, so that some confusions have been clarified.

However, I have still some additional comments, which are needed to be solved before publication.

Response to the Editor's Comments:

1. Comments/ errors: (lines in the revised version of MS)

Response: Sorry for our mistake. We have carefully checked the lines in this version of the revised manuscript.

2. Line 53: …wood burning was the most important….

Response: Sorry for our mistake. We have changed "were" to "was" according to your suggestion in Line 54, modified as shown below:

"Frka et al. (2018) found that wood burning was the most important source of humic-like substances (HULIS) in the aerosol accumulation mode (from ~0.1 to ~2 μm) during the autumn and winter;"

3. Line 55: Correct as: "…that HULIS in smaller particles was likely derived from local sources, while in larger particles from secondary organic aerosols (SOA)…"

Response: We are grateful for your valuable advice. According to your comment, the corresponding revision has been provided in Line 55-57 as follows:

"Jang et al. (2019) reported that HULIS in smaller particles was likely derived from local sources, while in larger particles from secondary organic aerosols (SOA) in the atmosphere,"

4. Line 101: Still not clear. “For each particle size, a quarter of each filter of all the collected samples in summer or winter season were mixed together. Do you mean that all quarters of filters of each size (summer/winter) were combined in one sample?

Response: Sorry for making such confusion. You are right that a quarter of all filters of each size (summer/winter) were mixed in a bottle and then ultrapure water was added to extract WSOC in this study. It has been revised in Line 102 and is now described as: “A quarter of all filters of each size (summer/winter) were mixed together in a bottle, extracted twice via ultrasonication in Milli-Q water for 15 min to achieve the extensive release of solubilized WSOC,”

5. Lines 190/191: “The average MAE_{365} values of particles in the size range of <0.26 μm, 0.44–0.77 μm, 1.40–2.50 μm, and 2.50–10.0 μm were 0.6937, 0.4656, 0.4610, 0.2426 $m^2\ g^{-1}$, respectively”. As I can see, these are the average values for summer and winter. Please, correct the sentence appropriately.

Response: Thanks for your comments. According to your suggestion, we have added the MAE_{365} values of different particle sizes in summer and winter in Line 190-193 as follows:

“The MAE_{365} values of particles in the size range of <0.26 μm, 0.44–0.77 μm, 1.40–2.50 μm, and 2.50–10.0 μm were 0.1258, 0.1321, 0.1014, and 0.1145 $m^2\ g^{-1}$ in summer, respectively, and 1.2615, 0.7991, 0.8206, and 0.3707 $m^2\ g^{-1}$ in winter, respectively, indicating that WSOC in smaller particles had stronger light absorption capabilities (Huang et al., 2022).”

6. Line 194: …the average AAE values were the highest in…

Response: Thanks for the helpful comments. According to your suggestion, we have revised this sentence in Line 195 as follows:

“The average AAE values were the highest in particle sizes of <0.26 μm,”

7. Line 193: ...nitrogen chromophores

Response: Thanks for your reminder. The corresponding revision has been provided

in Line 194 as follows:

"such as nitrogen chromophores (see Section 3.1.2),"

8. Line 255 (line 394): …was the highest in smaller particles (< 0.77 μm) and the lowest in larger particles…

Response: Thank you very much for your valuable suggestion. We have checked the whole text and made the following modifications in the revised manuscript:

Line 253-254: "the contribution of (strong) phenolic groups was the highest in smaller particles (< 0.77 μm) and the lowest in larger particles (1.40–2.50 μm)."

Line 282: "However, the MAE_{365} for particles < 0.26 μm in winter, exhibited the highest value at pH 4,"

Line 385-386: "whereas the contribution of phenolic groups was the highest in smaller particles (< 0.77 μm) and the lowest in larger particles (1.40–2.50 μm),"

9. Line 258: "…nitrophenols and their derivatives have been found to be possibly associated with the gas-phase oxidation of anthropogenic VOCs". Not only gas-phase, but may be also the result of aqueous-phase oxidation reactions.

Response: Thanks for your comments. We have revised it in Line 257-258 according to your suggestions, and the details are as follows:

"nitrophenols and their derivatives have been found to be possibly associated with the gas-phase oxidation of anthropogenic VOCs and aqueous-phase oxidation processes in polluted high-NO_x environments (Frka et al., 2022; Wang et al., 2019)."

10. Line 286: This sentence should be changed as: "The pH-dependent MAE_{365} suggests that under different pH conditions WSOC may have different impact on climate (i.e., climate impact would be enhanced as pH increases)."

Response: Thanks for your kind reminder. We have made the following modifications in Line 284-285 according to your suggestion:

"The pH-dependent MAE_{365} suggests that under different pH conditions WSOC may have a different impact on climate (i.e., climate impact would be enhanced as pH

increases) (Aiona et al., 2018)."

11. Line 290: … that the variations of the light absorption properties of BrC with pH were the result…

Response: Thank you very much for your suggestion. We have corrected it in Line 287-289 according to your suggestion:

"Phillips et al. (2017) found that the variations of the light absorption properties of BrC with pH were the result of structural changes in the nitro-aromatics and phenols."

12. Lines 292/293: "…to the greater content of aromatic species (e.g., nitrogenous aromatic species) in their WSOC. Better as: …to the higher content… (e.g. nitro-aromatic species) in WSOC.

Response: Thanks for your reminder. We have revised it in Line 290-291 as follows: "the strong dependence of light absorption properties on pH in smaller particles might be related to the higher content of aromatic species (e.g., nitro-aromatic species) in WSOC."

13. Lines 322/323 (and elsewhere): Please round the %! (e.g. 3.79%=3.8%).

Response: Thanks for your kind reminder. We have checked the whole text and rounded all the % in Lines 318-320 and elsewhere.

Line 271-273: "On average, the absorbance for particle sizes of < 0.26 μm, 0.44–0.77 μm, 1.40–2.50 μm, and 2.50–10.0 μm increased by 4.6 %, 1.3 %, 0.6 %, and 0.9 %, respectively, per unit pH increase in summer, and by 1.3 %, 0.5 %, 0.5 %, and 2.9 %, respectively, in winter."

Line 318-320: "On average, the FI/TOC of < 0.26 μm, 0.44–0.77 μm, 1.40–2.50 μm, and 2.50–10.0 μm decreased by 3.8 %, 3.5 %, 4.7 %, and 6.8 %, respectively, per unit pH increase in winter, which are significantly more than those (0.6 %, 1.7 %, 0.2 %, and 2.5 %, respectively) in summer."

14. Line 404: "as pH increases" can be deleted (as you already say "with increasing

pH").

Response: Thanks for your valuable comments. We have deleted "as pH increases" from this sentence in Line 395-396 as follows:

"The variation of both MAE_{365} and AQY of WSOC with increasing pH suggested the enhanced impact of WSOC on climate."