Measurement report: A new coupled method…

By Y. Qin et al.

General comments

In this paper, the authors report on the influence of pH and particle size on the chemical structures (i.e., functional groups, such as carboxylic, hydroxyl and phenolic) and optical properties (UV/fluorescence) of water-soluble organic carbon (WSOC) in aerosol particles, which were collected by MOUDI impactor during summer and winter. Their results provide information on WSOC sources and their aging processes in the atmosphere.

However, the content is mostly descriptive presenting the results with no deeper discussion, especially concerning atmospheric implications. In addition, the title sounds quite technical and the introduction is rather modest. Namely, there are many published articles on WSOC, also in size-segregated aerosols.

The manuscript could be of adequate atmospheric interest to merit publication in *Atmospheric Chemistry and Physics* as a *Measurement Report*, but after major revision, with addressing the following comments and/or questions.

Besides, I highly recommend the English language checking. I suggest also a separate list of abbreviations, which would be very helpful.

Data availability: Data have to be available in repository.

Specific comments

I suggest to change the title.

Some more references on WSOC in size-segregated aerosols should be involved in the Introduction.

Line 16: …source of these materials (not good word), source of WSOC constituents.

Line 32: "…ranging from 1 to 100 µm" (not really true). Important fraction represent particles below 1 µm. Please, check the definition.

Lines 45/46: Information on size-segregated WSOC: references are missing (e.g., Frka et al., Atmos. Environ. 2018)

Lines 74/75: Collection from June 2019 to August 2020, from 8 a.m. to 7 a.m. next day. This is one year. Can you explain how you got 82 samples only? In addition, if understood well, four size ranges have been considered (line 64), which means each sampling day 4 samples.

Line 80: Please, explain how the filters were combined for WSOC extraction.

Line 92: Why did you need to remove air from the WSOC solution?

Line 155: From the results in Table 1, I would not say that there is a big difference in $S_{275–295}$ for winter WSOC among all size ranges.

Line 156: Which results exactly show that? It is not clear.

Lines 154-172: This part is confused, and should be rewritten.
Line 158: It is written: "highlighting the relatively higher aromaticity of WSOC in small particles«;
on the other hand in line 156: "the molecular weight of WSOC was higher for particle sizes of 2.50–10.0 µm in summer and 1.40– 2.50 µm in winter",
and (line 166/167): "It has been documented that more aromaticity and larger molecular sizes of light absorbing substances tend to have higher MAE365, which is mainly derived…."

You concluded that the higher aromaticity is found in the smallest (below 0.26 µm), but on the other hand, higher molecular weight of WSOC was typical for bigger sizes.

Line 181: …"that aged WSOC undergone a growth process with increasing particle size". This statement is a bit strange. Please, correct.
Generally, with the process of growth the particle size increases.

Line 194: Both samples (which samples did you have in mind)?

Chapter 3.2.1: I suggest at least short concluding remarks from FTIR analyses.

Lines 209-210: This part can be moved in the Introduction.

Line 219: Not only phenol; so, it is better to say "aromatic compounds".

Lines 224/225: This is definitely not good enough / not a sufficient explanation. Just to remind, WSOC in winter (and summer) aerosols can be of primary and secondary source. Please, check some more references and add appropriate explanation.
For example, nitroaromatic compounds (phenolic like) can be largely formed via different reactions in gas or aqueous phase, and can be present in fine particles (below 1µm) in all seasons (See ref. e.g. Frka et al., Chemosphere 2022)

Line 232: Can you really say "monotonic"?

Line 236/237: Can you give some explanation?

Lines 243-246: Make two sentences.

Line 264: High pH values (above 7) are very rare for actual atmospheric conditions (clouds, fog, and aqueous aerosol), usually one can find more acidic conditions.

Lines 267/268: Please, give some explanation, why MAE_{365} for the smallest particles changed more.

Figure 6 is not necessary here, since the results are present also in Fig.7.

Line 281: The redshift and blueshift cannot be seen.

Lines 282-285: You speculate that –COOH and –OH groups influence the most on fluorescence behavior. What about aromatic groups (line 282)?

Line 303: From Fig. 9, I would not make such a conclusion. It can be seen that for both, HULIS1 and HULIS2, F_{max} decreased with pH very similarly (they had the same trend).

Lines 312-318: Not clear.

Lines 325/326: What can you say based on these results (what about aromatic compounds in winter WSOC samples)?

Chapter 4. Change the title as: "Summary and atmospheric implications"

Lines 338/339: As I said above, this is definitely not good enough. Please, correct.

Line 345: Please, see my comment above.

Figures: All figures/their subtitles are needed to be updated with missing information (see e.g. below for Fig.4).
Figure 1: Complete the information in the capture.
Figure 2: Complete the information in the capture.
Figure 3: Correct. In summer () and winter particles ()
Figure 4: Complete the information, for example as: Difference absorbance spectra (Δabsorbance) of WSOC in winter and summer particles of different sizes in the pH range……
Figure 5: Complete the information.
Figures 6, 7: Complete the information.

Technical corrections
Line 199: Correct the sentence.
Line 211:…in the range of 3.0-9.0.
Line 312: not materials/ use another word
Line 347: Delete "an indication of"