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Comment on acp-2022-84

Anonymous Referee #1

Referee comment on "Particle phase-state variability in the North Atlantic free troposphere during summertime is determined by atmospheric transport patterns and sources" by Zezhen Cheng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-84-RC1, 2022

This manuscript by Cheng et al. collected samples over three years at an interesting site (North Atlantic). They also used various measurement techniques (e.g., CCSEM-EDS and STXM-NEXAFS) for a significant number of samples as well as modeling and provided a unique conclusion regarding particle phases. Thus, I think this study will be an interesting contribution to our understanding of atmospheric aerosol particles.

Major comments.

1. I suggest including a discussion regarding the effect of relative humidity (RH) on the particle phase. Aerosol particle phases are sensitive to the RH when collected (e.g., Bateman et al. 2014 in the reference list). Inorganic aerosol particles can deliquesce, and organic particles can absorb water depending on RH, changing the shapes of sampled particles. The RH values should be obtained from an in-site measurement, if available, (not from a model result with a low spatial resolution) as the particle hygroscopicity is sensitive to the exact RH during the sampling. Although most particles should be in dry condition judging from Table S2, hysteresis phenomena may affect the particle hygroscopicity (e.g., Fig. S10). The current manuscript has a limited discussion regarding the ambient RH, and I suggest more discussion on RH effects for the particle phases. In addition, surface tension may also influence the height of the aspect ratio of sampled particles, and some discussion regarding surface tension may be useful.

2. The authors discuss the CO source contributions using the FLEXPART model. Although the model is acceptable and useful for CO, I wonder if it can be used to interpret the source of aerosol particles, especially for those with aging more than ten days. CO is gas and will not be removed from the atmosphere. On the other hand, a fraction of aerosol particles will be removed by mainly wet depositions during the transport with more than ten days (Table 1). Thus, it is not sure if the estimates of "contribution of source" in the table are valid for aerosol particles. Some explanation will be needed here.

3. Quality of Supporting information is a problem. The figures and captions include many errors, including the title (!), which is different from the manuscript. I wonder if the authors submitted the correct one or a draft version.

Specific comments.

4. Line 158. "an environmental SEM (ESEM) equipped with a FEI Quanta digital field emission gun, operated at 20 kV" and line 213 " Environmental Scanning Electron Microscopy (ESEM, Quanta 3D, Thermo Fisher)"

Are they different ESEM or the same one? The ESEM in line 158 is used for the CCSEM-EDS? It isn't very clear, and please specify them clearly.

5. Line 193 "inorganic components (In)"

In, IN, and "inorganics" are inconsistently used. For example, In is in line 207, "inorganics" is used in line 209, and IN is in line 324. In addition, "In" is confusing as it is like In (preposition).

6. Line 296-297. "Our particles are internally mixed based on tilted transmission electron microscopy (TEM, the titled angle was 70°) (Fig. S8)."

Please explain how to see Fig. S8, i.e., how the TEM image indicates internally mixed particles. Same for the description in line 328

7. Line 317-319. "Sulfate (CNOS and sea salt with sulfate) particles are also very abundant in all samples (~18 to 34 %), suggesting that these particles were involved in cloud processing (Ervens et al., 2011; Kim et al., 2019; Lee et al., 2011, 2012; Zhou et al., 2019)."

I am not sure why they were involved in cloud processing. Sulfate can originate from various processes. Does it mean organosulfates (CNOS)??

8. Line 324-325. states of OC (green), IN (blue), and EC (red) found in S3-3 and S4-2, which are (a) organic particle (green), (b) EC core (red) and coated by OC (green), (c) internally mixed EC (red) and In (cyan) coated by OC (green), and (d) In (cyan) coated by OC (green).

Both "cyan" and "blue" are used for In. I think it should be blue or IN and In are different??

9. Line 373-375. "These results suggest that apart from environmental factors, the inorganic components, the molecular weight of organic compounds, and the O/C ratio (or aging time) all affect the phase state of internally mixed particles."

They are true at specific RH values. For example, < RH 80%, ammonium sulfate is solid (crystal), and > RH 80%, they become liquid (deliquesce). These factors change the specific RH % that changes the particle phase state. Although it says "apart from environmental factors", some words about RH will be useful. Please see my comment 1.

10. Line 409-410. "Typically, particles with the same area equivalent diameter but higher TCA are more viscous (more solid-like) since they are less flat in shape (Fraund et al., 2020; Tomlin et al., 2020)."

The particle height may be also influenced by its surface tension if they are liquid. Please see my comment 1.

11. Figure 1. Please indicate what are the color indicate and what are the boxes and numbers.

12. Figure 2. These "solid black cycles" (circle?) are difficult to see with dark blue background.

13. Figure 3. Although I can imagine what the inserted normalized number fractions with size distributions in the upper right of each panel mean, it is better to have some explanation, especially the meanings of Y-axes.

14. Figure 4. Please indicate which samples were used for each panel.

15. Figure 5. Is panel (b) SA1 or SA2?

Table 1 indicates that 29.8% of SA1 particles are solid. Although I see SA1 includes relatively more semisolid particles, I cannot see solid particles. Could you indicate some examples of solid particles in the SEM images using ambient samples?

I also suggest adding RH values when collected for these samples.

16. Figure 6. In panel (a), there are 3 or 4 solid particles in SA2, but the solid particle % in SA2 is 0.0 in Table 1. Are they correct?

Supplementary information

I do not think I could indicate all errors. Please check the data carefully (or maybe it is a wrong file?).

17. The title is different from the main text.

18. Line 21. "where Tg,w is equal to 136 K, is the Tg for pure water,"

Tgw is 136K, correct?

" is the Tg for pure water " is correct?

19. Equation S3. C_real=(123.2±1.4)−(4.738±0.214)log(H)−(1.186±0.02)C_measured.

This equation indicates that less measured C atomic percentages yield a high "real" C percentage. I.e., if a particle includes no measured carbon percent (0%), it will have ~100 % of real C percent (by assuming H = 1). Although I do not have a way to check the accuracy, it is difficult to believe the result without more explanation. The calculation may influence the results in Figure S2, in which a fraction of particles consists of only C (no O nor other elements).

The equation S4 is also questionable. How can O=0%, which is seen in Fig. S2, be achieved?

O_real=(13.68±0.18)−(0.3413±0.0636)log(H)+(0.2579 ± 0.0072)O_measured (S4)

20. Line 49-51. "Since the particles are spheric, the measured area equivalent diameter (μm) is approximately equal to the height of particles. Therefore, when applying the correction function on our CCSEM-EDX data, we need to estimate the H by dividing the longest diameter retrieved from CCSEM-EDX measurement by the aspect ratio retrieved from tilted images (see Sect. 3.3.2). "

Do you have all aspect ratio data for all EDS measured particles? I think the aspect ratio was measured using ESEM, and the EDS was by CCSEM-EDS.

21. Table S1. Are there CCSEM data that can be listed for these samples?

22. Figure S2. These data, especially for C, look different between those from SA1 to S6 and those from S3-1 to S4-4 (different sampling periods). Are there any technical differences?

Potassium (K) may be used for a biomass-burning tracer. Have you checked it?

23. Figure S3. If you go to "No" and "No," you will find a question "Al+Si+Fe+Fe>Na", where you have double Fe.

24. Figure S4. Panel (a). There is "S-2," but it should be "S3-2." Y-axis should have "100" instead of "00". The caption should be "June" instead of "Jun."

25. Figure S5. The caption indicates from (a) to (i), whereas the panels are from (a) to (h).

26. Figure S7. "Jun" should be "June." Panel (a) and (b) is upside down. The legend in the panel (a, bottom) is overlapped with the plot.

27. Figure S8. Please indicate where we should see. Please see my comment 6.

28. Figure S9. The colors in OCInEC and In are nearly the same and cannot be distinguished. For example, in panel (f), it is difficult to identify if the light blue is OCInEC or In.

29. Figure S10. "Mean ambient temperature (blue)"

In the caption, the temperature is "blue," but in the legend, it is green. Same for Tg,org.

"(g) S3-2, (g) S4-3, (h) S4-4, and (i) S4-54. " There are two (g) in the caption. (i) should be S4-5 but no (i) in the panel (!!).

"uncertainties in RH (See SI). " Which SI should we see. we are now in SI.

30. References. The reference style is different from that of ACP.

31. Line 134 "Zieger, P. and Va, O" Please check the authors' name.