

We want to thank the reviewers for their comments. Addressing those comments has improved the quality of the manuscript. Below, we list each reviewer's comment (regular font), followed by our response (indented, **bold font**), followed by corresponding changes in the revised manuscript (indented, blue font). RL represents the line number in the revised version.

Anonymous Referee #2

The authors described the phase states of aerosol particles collected in the North Atlantic FT and tried to explore the transport patterns of the aerosol particles. Such research topic is interesting for the atmospheric communities, and also the scope of the research is suitable in ACP journal. However, after carefully reviewing this manuscript, the evidence are rather weak to support the results, and conclusion is too generalized. In addition, many errors in the text, figures, Tables, and SI can be founded.

We appreciate the constructive feedback from the reviewer. We attempted to streamline some conclusions and noted the study's limitations. Below are our responses to each comment:

Major comments:

1. During the laboratory experiments for the phase determination, at which relative humidity and temperature the ESEM did the authors perform? This should be clearly stated in the manuscript. The main issue is that how the authors can conclude the phase states of the aerosol particles if the relative humidity and temperature during the experiments were different compared to the field measurement periods? The phase states of aerosols are temperature- and relative humidity-dependent, and thus it didn't convince me whether the conclusion is still valid or not. This should be clearly mentioned through the manuscript. The authors should also show the ambient RH and temperature at the monitoring site in a figure and table.

We appreciate that reviewer pointed this out. Section 2.3 RL167 mentioned that “Ambient particle samples were analyzed with ESEM at 293 K and under vacuum conditions ($\sim 2 \times 10^{-6}$ Torr)”. These conditions are not representative of the ambient atmosphere. Our measurements capture the phase state of particles at the time of sample collection. We agree that, in principle, changes in temperature and humidity in the ESEM chamber (under vacuum conditions, the RH inside the chamber is close to 0%) could affect the phase state of an airborne particle. However, our inference of the particle's phase state at the time of collection is based on the shape the particle acquires at impaction on the substrate, which unlikely would change significantly within the ESEM chamber due to adhesion forces between the particle and the substrate. This is a caveat of this method, which has been reported in previous studies. (e.g., Cheng et al., 2021; Lata et al., 2021; Wang et al., 2016); however, we believe that these results still provide useful information about the phase state of the particles in the atmosphere. Future studies should focus on determining the uncertainties introduced by RH-dependent phase states. To make this point clear, we add the following sentences in L167:

RL167-174: “Ambient particle samples were analyzed with ESEM at 293 K, under vacuum conditions ($\sim 2 \times 10^{-6}$ Torr) and therefore at RH values near zero, which might lead to losses of volatile and semivolatile materials. Moreover, the temperature and RH inside the ESEM chamber differed from those at the OMP during sample collections (Fig. S2). RH and T affect the phase state of airborne particles; however, our inference of the particle's phase state at the time of

collection is based on the shape the particle acquires at impaction on the substrate, which unlikely would change significantly within the ESEM chamber due to adhesion forces between the particle and the substrate. These limitations need to be considered when interpreting our results.”

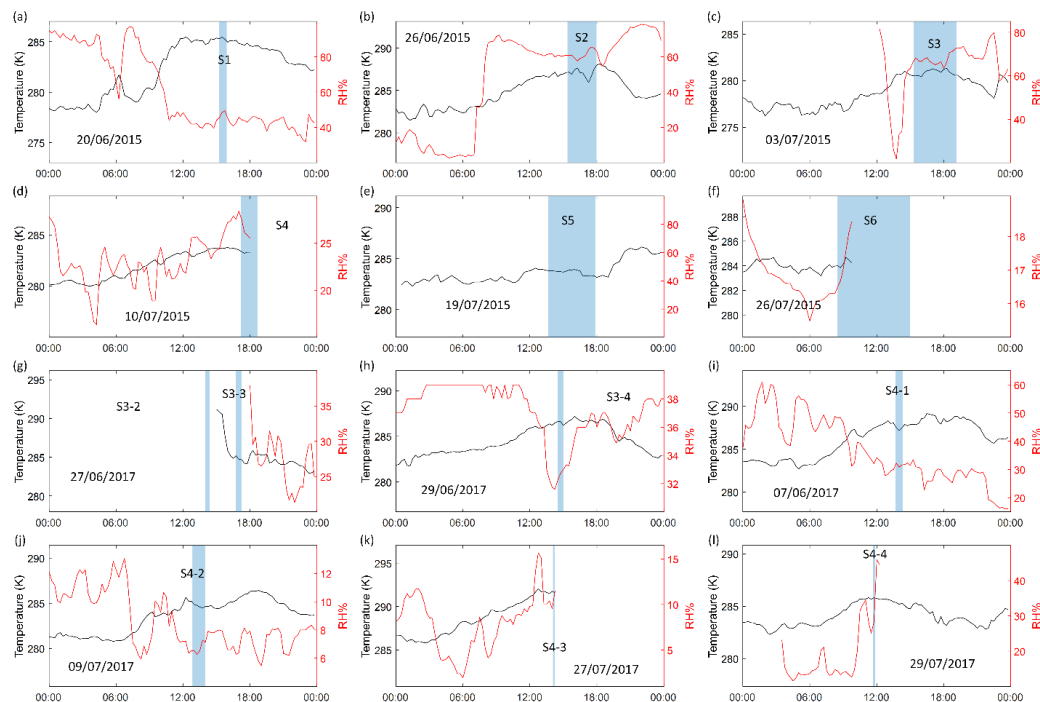


Figure S2. Hourly variation of temperature and relative humidity for available days. Shaded areas represent the sample collection periods.

- Regarding the technique of the tilted aspect ratios to determine the phase state of aerosols, I am confusing this technique is reliable for aerosols consisting of mixtures of organic materials and inorganic compounds. The authors should validate and carefully described the evaluation of the results with comparison to previous phase studies using well-known mixtures or commercial standards comprising organic and inorganics. I cannot find such validation from Cheng et al. 2021.

We appreciate that reviewer for pointing this out. The technique of tilted aspect ratio has been used to study ambient particles in previous studies (e.g., Cheng et al., 2021; Fraund et al., 2020; Lata et al., 2021; Sharma et al., 2018; Tomlin et al., 2020; Veghte et al., 2017; Wang et al., 2016). We agree with the reviewer that the aspect ratio thresholds used to define the phase state were based on standard organic material, but we showed that the technique was also applicable to field-collected samples in Cheng et al., 2021. In addition, we also utilize the STXM/NEXAFS measurements to determine the phase state, which was also applied in previous studies (O’Brien et al., 2014; Tomlin et al., 2020). We agree with the reviewer that mixtures of organic and inorganic standards would be useful to refine those thresholds further. However, we think these results are still valuable to assess the phase state of individual submicron size particles, and future studies should focus on measurements of phase state from tilted view imaging. Thus, we plan to directly measure the viscosity from standard organic-inorganic mixtures in a future study.

To clarify the limitation of the study, we added the following sentences in the revised manuscript:

RL549-555: “This study assesses the phase state of internally mixed FT particles at the time of sample collection, and highlights the importance of accounting for inorganic inclusions to evaluate the phase state of internally mixed particles. Our results might not fully represent the phase state of FT particles during transport due to differences in ambient temperature and RH. Moreover, the aspect ratio thresholds used to determine the particles’ phase states are based on limited standards. Future studies should focus on improving the aspect ratio thresholds by using more standards with known viscosities and determining the viscosity of internally mixed individual particles as a function of temperature and RH.”

3. Figures and SI should be revised (see also below). Moreover, all figures in SI should be mentioned in the main text.

We thank the reviewer for this comment. We have made corrections in the revised version.

Minor comments:

4. Page 5 line 136: The author should provide more details about stored conditions by mentioning temperature. Furthermore, the authors have to mention the stored period before the experiment due to evaporation issue.

In the main manuscript RL138-139, we mentioned that “Samples were stored at ambient condition and wrapped in Al foil immediately and kept in zip lock bags after collection to avoid exposure to light and air and minimize potential modification and oxidation in the air.” Moreover, Pico 2017 samples were analyzed in the same year as soon as we received them from OMP. We agree that some sample modifications might occur during storage, but this is a limitation for any offline analysis of field-collected samples. We also underline that the site is quite challenging to access, making prompt sample analysis much more challenging than other sites; however, this aspect also makes the samples particularly unique and valuable. Thus, we add the following sentence in L137 in the revised manuscript:

RL141-145: “Samples were placed in dedicated storage boxes wrapped in Al foil and kept in zip lock bags immediately after collection to avoid exposure to light and outside air. The samples were then stored at ambient conditions to reduce the chances of modifications and oxidation that might have partially intercurrent. This is a limitation of any offline analysis of field samples. We underline that the site is quite difficult to access; therefore, samples were delivered and analyzed as soon as it was feasible (less than one year after collection).”

5. The authors should provide details about the particle regeneration in the experimental section if it regenerated from the collected samples.

We did not regenerate any particles in this study. Particles were collected on TEM grids (carbon-B film), and analyses were performed directly on those grids.

6. There are too many academic terms in the manuscript and it is suggested to add a table to summarize all acronyms and full names. The authors repeatedly used a similar abbreviation for the OC component with different names such as Organic (OC) (Page 7 line 192), and organic carbonaceous (OC) (Page 6 line 170). Abbreviation similarity should be consistent without repetition.

We thank the reviewer for this great suggestion. We have added a Table of Acronyms and Abbreviations (Table 2) in the main manuscript and revised L170 as below:

RL183-185: “Based on their element percentage, each particle in Pico 2017 can be classified as organic (OC), carbonaceous with nitrogen (CNO), carbonaceous with sulfate (CNOS), sea salt (Na-rich), sea salt with sulfate (Na-rich with S), dust (Al, Si, Ca, Fe), dust with sulfate (Al, Si, Ca, Fe, S), and others (see Fig. S4).”

7. Page 4 line 119: The author mentioned “This study focuses on detailed individual particle analysis on Pico 2017”. In addition, on page 6 line 172, the authors mentioned “CCSEM-EDX based particle classification for Pico 2014 can be found in Lata et al., 2021, and that for Pico 2015 will be discussed in our future work”. However, some data relevant to the phase state for the 2014 and 2015 shown in Fig. 5. Also, total carbon absorption (TCA) data showed in fig 6 for Pico 2014, and Pico 2015. It makes confusion to the readers regarding which data Pico 2014, Pico 2015, or Pico 2017 is exactly discussed in this manuscript. To avoid more confusion author has to focus more on Pico 2017 data or the data relevant to Pico 2014 and Pico 2015 should move to SI.

We agree with the reviewer that all the data might be confusing for the readers without proper description. We tried to address this comment in our revised manuscript. The CCSEM-EDX analysis and FLEXPART simulation for Pico 2014 has been discussed in detail in Lata et al., 2021, and these for Pico 2015 will be discussed in future work. The main focus of this manuscript is the phase state of particles in the North Atlantic free troposphere during summertime based on samples collected over three different years. Thus, the tilted view imaging (Fig. 5) from ESEM and TCA analysis from STXM (Fig. 6) are crucial. However, to help our discussion of the association between chemical composition and source contributions with phase state, we decided to add short summaries of tilted view imaging analysis and TCA analysis from Pico 2014 and 2015. Therefore, we believe keeping Fig. 5 and Fig. 6 with data from all three samples can help generalize the findings and help the reader understand our main findings. We modified the text as follows in the revised manuscript RL116-122.

RL117-123: “In this study, we present an overview of the phase state of individual FT atmospheric aerosol particles collected at OMP over three different years, which are July 2014 (Pico 2014), June and July 2015 (Pico 2015), and 2017 (Pico 2017). Analysis of samples from three years using tilted view Environmental Scanning Electron Microscope imaging and scanning transmission X-ray microscopy with near-edge X-ray absorption fine structure spectroscopy (STXM-NEXAFS) are reported to study the phase state of individual particles. The chemical composition and phase state of individual particles for Pico 2014 have been reported in a previous study (Lata et al., 2021). The chemical composition of individual particles for Pico 2015 will be discussed

in future work. This study focuses on detailed individual particle analysis of the Pico 2017 samples.”

8. Page 8: In the result and discussion section, the description of Fig. 1 looks confusing and keeps the reader browsing to keep up with the text. The text is littered with redundant statements in parentheses that re-state what has just been explained. Please specify them clearly.

We appreciate this comment from the reviewer and are sorry for the confusion. This study discussed eight FLAXPART retroplume analyses and the related CO tracer simulation results for the period the Pico 2017 samples were collected over, which were not discussed before. Some of the statements in parentheses have indeed been provided in Table 1, but we believe showing the numbers in the main body of the paper can help readers understand the contribution of CO from different sources, which are important for our discussion in Section 3.3.1. Nevertheless, we agree with the reviewer that it might be difficult for readers to keep up with the text. We attempted to reduce the redundant statements.

9. Page 10 lines 290-294: More careful and detailed description are needed for Fig. 2 by comparing it with the reported study because size distribution is a very important factor when defining the physicochemical properties of an ambient particle. Also, please add how you measured in Experimental.

Thanks for the comment. We agree with the reviewer that size distribution is a critical factor in understanding ambient particles' physicochemical properties. The detail of SMPS measurements has been described in (Siebert et al., 2021), which reports details on the Azores Stratocumulus Measurements of Radiation, Turbulence and Aerosols (ACORES) campaign in July 2017 that took place in the Azores, including activities at OMP. Therefore, based on the reviewer's suggestion, we revised L290-294 as below:

RL303-313: “Figure 2 shows the particle size distribution and the total particle concentration based on SMPS measurements at OMP, and CO tracer concentrations in the air masses that arrived at OMP as retrieved from FLAXPART simulations (5 July 2017 to 21 July 2017). Mobility diameter ranged from 30 nm to 500 nm, and the mode was around 60 ± 22 nm (Fig. 2a). The total particle concentration was around 279 ± 114 # cm^{-3} . The size range, size mode, and particle concentration were comparable to those found in previous studies for FT particles (10-1000 nm, <100 nm, 10^1 to 10^4 # cm^{-3} , respectively) (Igel et al., 2017; Rose et al., 2017; Sanchez et al., 2018; Schmeissner et al., 2011; Sun et al., 2021; Venzac et al., 2009; Zhao et al., 2020). Figure 2b shows that the total particle concentrations positively correlate with the CO tracer concentrations from July 5th to July 12th and from July 18th to 21st, suggesting the major sources of particles during these periods might be anthropogenic and wildfire emissions. On the other hand, particle concentrations between late July 12th and 17th were above 279 # cm^{-3} , while the CO tracer level was relatively low (<10 ppbv) compared to other days, which might indicate additional sources of particles (e.g., sea spray and dust).”

We also added more details regarding the SMPS measurements in the Experiment section:

RL145-147: “Moreover, from 05 July to 21 July 2017, we also deployed a Scanning Mobility Particle Sizer (SMPS, TROPOS, for details, see Wiedensohler et al., 2012) coupled with a silica gel diffusion dryer to monitor the dry particle size distribution (<40% RH) and the total particle concentration with 5 mins time resolution (Siebert et al., 2021).”

10. To make this manuscript understandable to the readers, I would like to suggest the authors move data relevant Pico 2014, Pico 2015 to the supporting information. It has been already published.

We thank the reviewer for this valid suggestion. Please see our response to comment 7.

11. The authors didn't describe clearly which samples were used for Fig. 4 which is relevant to STXM/NEXAFS spectra, Is that data relevant to Pico 2017? Even though there is no clear evidence in the description part (Page 11 line 323 to 329).

Thanks to the reviewer for this suggestion. These particles were from S3-3 and S4-2 from Pico 2017. These are representative particles to help readers understand typical STXM-NEXAFS spectra and maps for four types of particles. We have revised the manuscript:

RL344-347: “Figure 4 shows STXM-NEXAFS Carbon K-edge chemical speciation maps and spectra for four typical particle mixing states of OC (green), IN (cyan), 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).”

And:

Figure 4. Representative STXM-NEXAFS spectra of (a) organic particle (green), (b) EC core (red) coated by OC (green), (c) internally mixed elemental carbon (red), and IN (cyan) core coated by OC (green), and (d) IN (cyan) core coated by OC (green) from Pico 2017 S3-3 and S4-2 samples. White scale bars represent 500 nm.

12. Please clarify the captions of the SI.

We appreciate the reviewer for pointing this out. We have made the necessary changes to make the captions of the SI clearer. Please check the revised version.

13. The title should be revised based on the main findings.

We updated the title in the SI, and it is now the same as the main text. Our title is based on the main findings.