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 and RSL represent the line number in the revised main manuscript and SI, respectively.

Anonymous Referee #3

Cheng et al. made a comprehensive investigation on the sources, chemical composition, and phase state of long-range transported free tropospheric particles based on individual particle analysis using multi-modal micro-spectroscopy techniques. This study found that most particles were in the liquid state and highlighted the importance of considering the mixing state, emission source, and transport patterns of particles when estimating particle phase state in the free troposphere. Though the findings are expected, the observation data provide valuable information constraining the physiochemical properties of aerosols in the free troposphere which is important in assessing aerosol associated climate effects. I agree with the comments of the other two referees that this manuscript can be published only after a major revision as there are too many errors in the submitted version.

> **We appreciate the positive feedback and constructive criticism from the reviewer. We attempted to fix all the errors in the manuscript. The reviewer raised some critical points, which we believe were addressed in the revised version and strengthened the article. Below are our responses to each comment:**

Major comments:

1. **We want to thank the reviewer for these important comments. Please see our response to each of points below:**

   a. My major concern is on the Section 3.3.2, the phase state of particles during long-range transport. The authors mainly investigated the phase state of organic particles applying the temperature and RH along the air mass transport path, and found that organic particles would likely be solid in most of the times. As the particle viscosity depends significantly on RH as pointed by the other two referees, I doubt the meaningfulness investigating the phase state at each air mass path with a wide variation in RH as shown in Fig. S10.

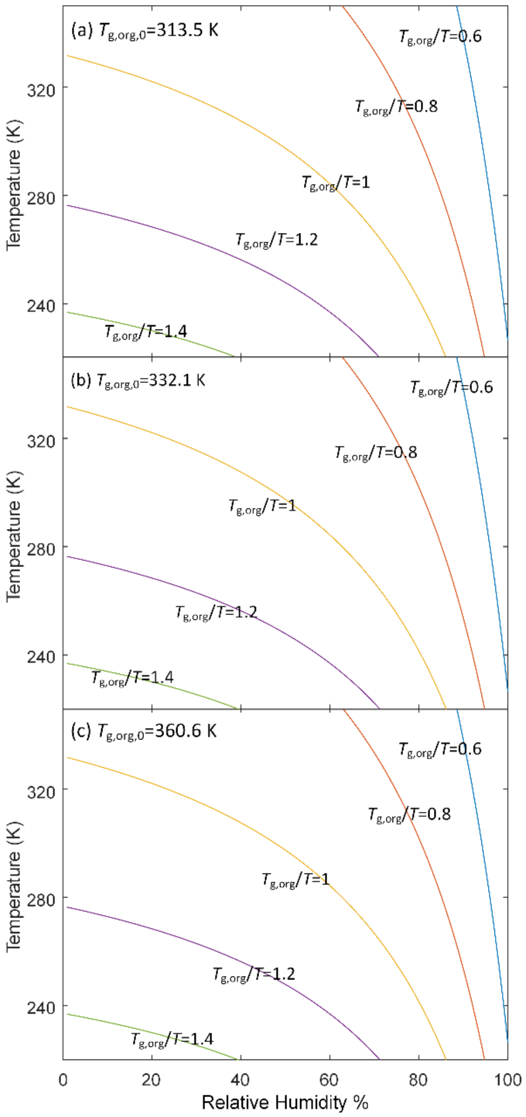
   > **We agree with the reviewer that temperature, relative humidity, and chemical composition of particles in the free troposphere are highly variable, and the phase state and viscosity of those particles depend on these factors, leading to large uncertainties. It would be ideal to measure the ambient conditions of air masses and phase state and viscosity of FT particles at multiple points over their transport path, but that is not feasible. Therefore, we resort to models to predict RH and T during transport. As shown in Fig. S6 in the revised paper, the vertical dispersion of air masses increases significantly after more than 5 upwind days, leading to significant variations in retrieved temperature and RH values. To minimize the induced uncertainties in the glass temperature calculations, we only predicted $T_{g,org}$ up to 5 days before the air mass reached OMP. We acknowledge these limitations in the current version of the manuscript; however, we still believe that the analysis provides valuable information to understand the evolution of the phase state of FT organic particles. We note that a similar approach was also adopted in a previous publication (i.e., Schum et al., 2018 ACP). To make this clear, we add the following sentences in RL481-483:**

RL484-486: **“**The estimates only up to 5 days before air masses arrived at OMP to avoid increased uncertainties associated with possible meteorological conditions due to the spread of the air masses (see Fig. S6).”

b. I suggest adding a figure showing the variation of predicted viscosity with RH and T (similar to the figures in (Li and Shiraiwa 2019, Petters, Kreidenweis et al. 2019)) and investigating the phase state at free troposphere-relevant conditions.

As the reviewer suggested, we added Fig. S12 in SI and a sentence in RL484-485.

RL486-487: “Predicted $T_{g.org}$ to T ratio as a function of temperature and relative humidity are shown in Fig. S12.”



“Figure S12. $T_{g,org}/T$ ratio as a function of temperature and relative humidity for organic particles transport in FT by using (a) minimum,

(b) median, and (c) maximum dry glass transition temperatures of organics as reported in Schum et al., 2018 (313.5 K, 332.1 K, and 360.65 K, respectively)."

c. In addition, the authors applied a single value for the dry glass transition temperature, which, however, would be changed due to the change in the chemical composition during the long range transport.

As mentioned earlier, we fully agree with the reviewer that dry glass transition temperatures will differ for different chemical compositions, which might change during the long-range transport. To study the sensitivity of the dry glass transition temperatures to different chemical compositions, we used the minimum and maximum dry glass transition temperatures of organic particles as reported in Schum et al., 2018 (360.65K and 313.46K, respectively). The ranges of glass transition temperatures are shown as the shaded areas of predict $T_{g,org}$ in Fig. S11 in the revision, resulting from variations in temperature, RH, and chemical composition.

d. Finally, could the authors add some discussion that based on the inorganic component types you have observed, how you expect the phase state variation of inorganic components during the long range transport at free tropospheric RH and T? It would be helpful supporting the implication what you wrote in the Conclusion section that the particles in the FT probably remain liquid.

We thank the reviewer for pointing this out. In Sect. 3.2, we have shown that our samples were internally mixed with hygroscopic salts such as sea salt and sulfate. Therefore, we expected that the presence of these inorganic inclusions would reduce the viscosity of FT particles at the RH and T values encountered in the FT during transport. This has been discussed in the last paragraph in Sect. 3.3.2. To make this clearer to the readers, we added the following sentence in the revised manuscript:

> RL509-512: "Thus, our results suggest that estimating the phase state of particles without considering the mixing state of FT particles might not accurately predict their viscosity and T_g because the presence of hygroscopic inorganic inclusions (e.g., sea salt and sulfate) can reduce the viscosity of FT particles at the RH and T values encountered in the FT during transport."

We further added one sentence in the conclusion section:

> RL541-542: "Moreover, the fraction and chemical composition of inorganic inclusions may further influence the phase state variations."

2. Mixing state plays an important role in the phase state of ambient particles; however, the authors did not mention other factors that may impact phase state significantly. Besides the influences of surface tension on aspect ratio and thus the prediction of phase state mentioned by Referee #1, the influence of particle size should be considered and discussed as well. Several studies have found that the size of particles influence the viscosity (Cheng, Su et al. 2015, Petters and Kasparoglu

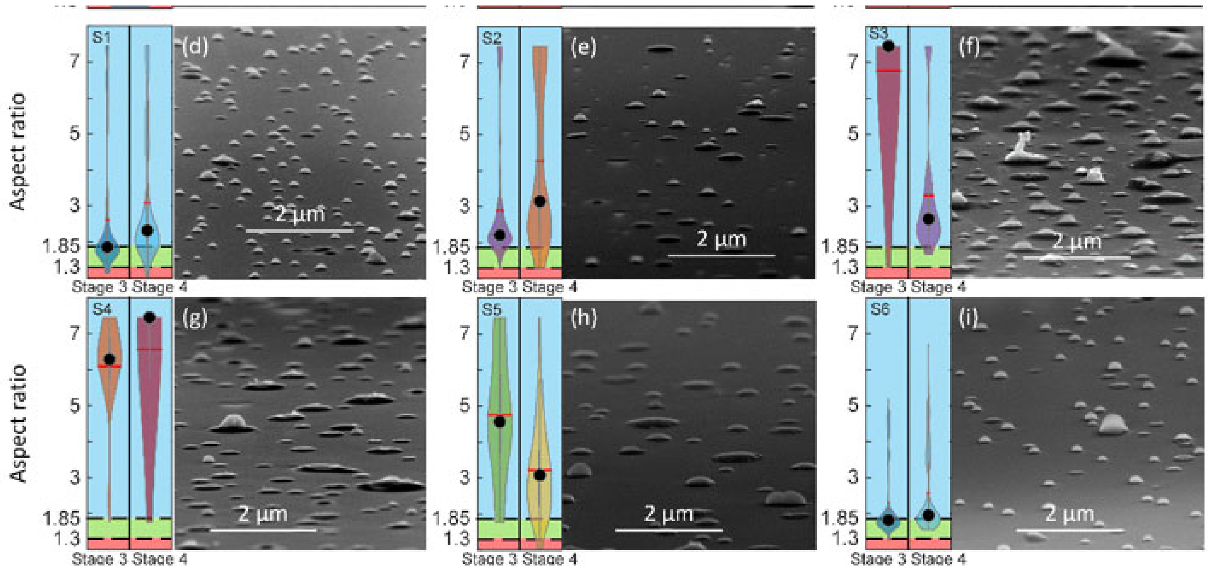
2020). Did you see the difference in the phase state between the particles collected on the 3rd and 4th states of the impactor? Would the change of particle size affect the phase state during the long range transport? Secondly, the authors only mentioned the inorganic components could decrease the viscosity of internally mixed particles. They missed a recent study showing that increasing inorganic fraction can increase aerosol viscosity through cooperative ion-molecule interactions (Richards, Trobaugh et al. 2020).

We want first to thank the reviewer for these very constructive comments. Please see our responses to your questions below:

a. Did you see the difference in the phase state between the particles collected on the 3rd and 4th states of the impactor? Would the change of particle size affect the phase state during the long range transport?

We did look at the size dependence of viscosity. In this study, SA1-SA3 and S3-1-S3-4 were collected on stage 3, and S4-1 to S4-4 were collected on stage 4. Since these samples were collected during different times, we are reluctant to draw any firm conclusions about the dependence of viscosity and phase state on particle size differences based only on these samples. On the other hand, samples S1-S6 were collected on stage 3 and stage 4 simultaneously, and we revised Fig. 5 (the figure below shows the revised portion of Fig. 5) to show the stage difference of aspect ratio distribution for these samples. As shown in revised Fig. 5, aspect ratios are different between stage 3 and stage 4. The particles collected on stage 3 for samples S1, S2, S4, and S6 have a higher fraction of more viscous particles than those collected on stage 4. However, particles collected on stage 3 for samples S3 and S5 have lower fractions of more viscose particles than those collected on stage 4. Therefore, we do not see a clear trend in the size dependence of the particle viscosity. However, we agree with the reviewer that this is worth further study. Hence, we added the following discussion in the revised manuscript:

RL413-421: “Moreover, Cheng et al., 2015, Petters and Kasparoglu, 2020, and Kaluarachchi et al., 2022 have shown that particle size also affects the particles' viscosity. This appears to be the case for some samples when comparing the aspect ratio distribution for the Pico 2015 particles collected on stage 3 (left violin plots, 50 % cut-off size is >0.15 μm) with those from stage 4 (right violin plots, 50 % cut-off size is >0.05 μm) in Fig. 5d to 5i. For samples S1, S2, S4, and S6, particles from stage 3 have lower mode and mean aspect ratio than those from stage 4, indicating that larger particles have higher fractions of more viscous particles than smaller particles. However, the aspect ratio distributions for particles collected on stage 3 in samples S3 and S5 have higher modes and mean values than those on stage 4, suggesting a higher fraction of less viscous particles in S3 and S5 on stage 3. Due to these inconsistent observations and the limited number of samples, we cannot draw clear conclusions regarding the size dependence of particle viscosity; this important aspect should be the objective of future studies."



b. Secondly, the authors only mentioned the inorganic components could decrease the viscosity of internally mixed particles. They missed a recent study showing that increasing inorganic fraction can increase aerosol viscosity through cooperative ion-molecule interactions (Richards, Trobaugh et al. 2020).

Thanks for pointing out this paper. Richards et al., 2020 show that divalent ions (e.g., Mg^{2+} and Ca^{2+}) can enhance the viscosities of organics through ion-molecule interactions. Unfortunately, we cannot identify the chemical formula involving these divalent ions with our technique. However, in our samples, the total percentage of these elements is very low (~0.17±0.34 %), while Na (~0.61±1.0 %) and sulfate

(~0.48±0.40 %) are more abundant. Thus, the effect of ion-molecule interactions might not be critical for the particles investigated in our study. However, we agree that this is an important mechanism that should be discussed in the manuscript. To make this clear, we revised the manuscript and added the discussion below:

> RL379-382: "Besides, Richards et al., 2020 have reported that divalent ions (e.g., Mg^{2+} and Ca^{2+}) can increase aerosol viscosity due to ion-molecule interactions. Although our analytical technique cannot identify the chemical formula involving these divalent ions, this phenomenon might not be critical for our samples because we found only minor fractions of Mg and Ca."

Specific comments:

Manuscript:

1. I recall the comments by the other two referees that the RH in the ESEM should be clearly pointed out as the particle phase state depends significantly on RH.

   We thank the reviewer for making these valid comments. Please refer to our response to reviewer 2, major comment 1:

   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 (~2×10^{-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 sentence in L161:

   > RL167-174: "Ambient particle samples were analyzed with ESEM at 293 K, under vacuum conditions (~2×10-6 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."

Moreover, as the reviewer suggested, we added the average temperature and RH at OMP during the sample periods to Table S1 and S2 and plotted the hourly variation of temperature and RH in Fig. S2 in revision:

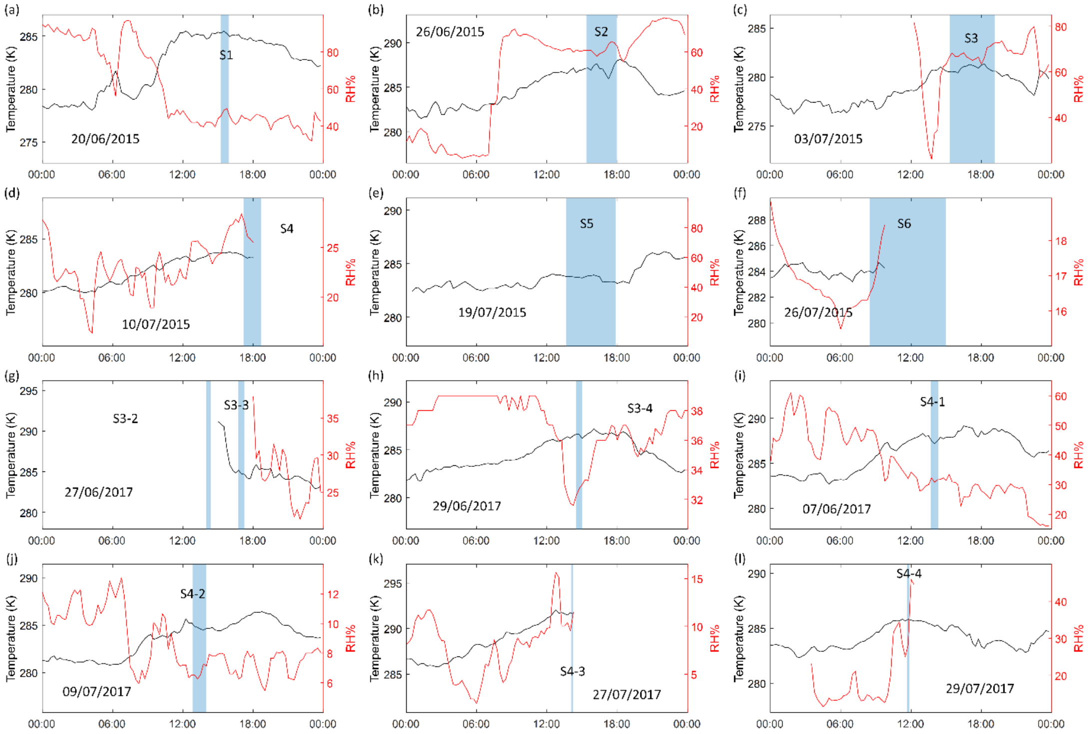


Figure S2. Hourly variation of temperature and relative humidity at OMP for available days. Shaded areas represent the sample collection periods.

2. Give the full name of "SEM" at Line 68 instead of at Line 71. Are SEM and ESEM the same?

We are sorry that we did not make this clear. We used the same ESEM for both CCSEM-EDX and tilted imaging experiments. We revised line 158 and line 213 as below:

RL167-168: "We utilized an environmental SEM (ESEM, Quanta 3D, Thermo Fisher) equipped with a FEI Quanta digital field emission gun, operated at 20 kV and 480 pA."

RL226-228: "We utilized tilted view imaging combined with the ESEM to estimate the phase state of particles based on their shapes. For each sample, we evaluated more than 150 randomly selected particles. Moreover, tilted view imaging and CCSEM-EDX experiments were performed independently."

3. Line 125. Change "Experimental" to Experiments.

Change has been made.

4. Line 177-178. “87 for S3 and 37 for S5 for Pico 2015, and 142 and 171 particles for S3-3 and S4-2 for Pico 2017”. These data are not same as those in Table S2 and Table S3. Please check which are correct.

    Thanks for this comment. We have revised the sentence as below:

    > RL188-192: “Due to beamline time constraints for STXM analysis, we focused only on selected samples and a limited number of selected particles (653 for SA1, 208 for SA2, and 425 for SA3 for Pico 2014, 86 for S3 and 37 for S5 for Pico 2015, and 140 and 166 particles for S3-3 and S4-2 for Pico 2017).”

5. Line 190, I don’t understand what TCA is proportional to?

    We apologize for the confusion. TCA is proportional to the particle thickness based on Eqn. 1 and 2. We modified the sentence as follows:

    > RL203-204: “Thus, TCA is proportional to the particle thickness, and it can be used as an indicator for particle thickness (O’Brien et al., 2014; Tomlin et al., 2020).”

6. Line 245. Explain how you determined the air mass source is wildfire from “CO source contributions”.

    PMO is usually dominated by outflows of anthropogenic emissions from North America and is occasionally affected by wildfire events. Wildfire events at PMO can contribute as much as anthropogenic emissions when they occur, enhancing CO by ~25 ppbv upon the North Atlantic background on average. In terms of aerosol composition, wildfire events have a clearer signature, such as enhanced BC, so we consider any samples with over 20% of FLEXPART CO from biomass burning would have a clear impact on the aerosol composition and properties of collected samples.

7. Line 277-281, the data described for SA1, SA2 and SA3 are different from the corresponding data in Table 1.

    We have revised the sentence as below:

    > RL290-295: “Based on the CO tracer analysis, the major CO sources for SA1 were anthropogenic emissions in North America (~49 %), anthropogenic emissions in South America (~8 %), and wildfires in North America (~19 %). For SA2, the major CO sources were North American anthropogenic emissions (~42 %), African anthropogenic emissions (~16 %), and North American wildfires (~31 %). For SA3, anthropogenic (~49 %) and wildfire (~49 %) emissions in North America were the two major CO contributors (Lata et al., 2021).”

8. Line 284-286, “and S1, S3, and S6 were influenced by both anthropogenic and wildfire CO emissions in North America (~56 %, ~79 %, ~40 %, and ~59 % for anthropogenic CO source,

and ~42 %, ~19 %, ~53 %, and ~25 % for wildfire CO sources, respectively).” Check the values (there are four values for three samples).

We have revised the sentence as below:

> RL296-299: “Based on the CO tracer simulations (Fig. S8), the major source of CO for sample S2 was anthropogenic emissions in North America (~84 %), and S1, S3, S5, and S6 were influenced by both anthropogenic and wildfires CO emissions in North America (~56 %, ~79 %, ~38 %, and ~59 % for anthropogenic CO sources, and ~42 %, ~19 %, ~53 %, and ~25 % for wildfires CO sources, respectively).”

9. Line 289. Change “Chemical-resolved” to “Chemically-resolved”.

Change has been made.

10. Line 292. Change “>400 particles cm-3” to “>400 particles cm-3 ”

We made the change as suggested by the reviewer.

11. Line 296. “Our particles are internally mixed based on tilted transmission electron microscopy (TEM, the titled angle was 70°) (Fig. S8).”, Line 328. “This observation is consistent with their STXM images and tilted TEM images (Fig. S8)”. Give a more detailed explanation how an internal mixing state is determined? Line 297. Change “titled angle” to “tilted angle”.

Thanks for bringing up this point. We can see internally mixed EC and inorganic inclusions coated by organics in representative tilted TEM images (Fig. S9 in revision) and STXM images of individual particles (Fig. 4). To make this clearer, we have revised the manuscript as below:

> RL316-317: “Tilted transmission electron microscopy (tilted angle 70°) images show that inorganic inclusions (e.g., sea salt, nitrate, sulfate, dust) are internally mixed and coated by organics (Fig. S9).”

And:

> RL349-352: “Moreover, STXM images (see Fig. 4) indicate that particles are internally mixed and coated by organic species, suggesting our samples might be highly aged during transport in the FT (China et al., 2015; Motos et al., 2020). This observation is consistent with our tilted TEM images showing that EC and IN inclusions were internally mixed with organics (Fig. S9).”

12. Line 298. “Fig. 2(b to i) show” should be Fig. 3(b to i). g

We have corrected it:

> RL317-319: “Figure 3a shows the average number fraction of different particle types in each sample, and Fig. 3b to 3i show chemically-specific normalized particle size distributions.”

13. Line 304. “area equivalence diameter” . Do you mean “area equivalent diameter”?

    We have revised the sentence:

    > RL322-325: “Sea salt with sulfate particles with area equivalent diameters greater than 0.6 μm have been shown to be a product of aqueous phase processing (i.e., fog and cloud processing) (Ervens et al., 2011; Kim et al., 2019; Lee et al., 2011, 2012; Zhou et al., 2019), and those with area equivalent diameter less than 0.6 μm might have been generated from marine sources (Sorooshian et al., 2007; Yu et al., 2005).”

14. Line 306. The values of 79.6% and 1.1% did not match the values in Table S2.

    We have corrected the numbers.

    > RL325-327: “Sea salt and sea salt with sulfate particles dominated (~28.2 % and ~31.5 %, respectively) sample S3-1, with a smaller fraction of organic particles (OC and CNO, ~6.3 % and ~23.4 %, respectively) than in other samples.”

15. Line 323. “Figure 4 shows the STXM-NEXAFS Carbon K-edge chemical speciation maps and spectra of four typical particle mixing 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)”. Do “blue” and “cyan” both indicate the inorganics? DO “IN” and “In” both indicate the inorganics? And the description here is different from the caption of Figure 4.

    Sorry for the confusion. We used IN for inorganic components retrieved from STXM-NEXAFS spectroscopy and made changes accordingly in the revised manuscript. We also have corrected our manuscript and used cyan for IN in Fig. 4.

    > 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).”

    We also corrected the caption of Fig. 4:

    > **“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.”**

16. Line 331. “all samples” Check Figure S9 is for the results of all samples of only seven samples.

To make this sentence clearer, we revised it as below:

> RL354-355: "The particle chemically-resolved size distributions for seven samples (SA1-SA3, S3, S5, S3-3, and S4-2) analyzed with STXM-NEXAFS are shown in Fig. S10."

17. Line 332. "S3-3 and S4-4 samples" Do you actually mean S3-3 and S4-2 samples? I do not see S4-4 in Figure S9, and in Table S2, the sample analyzed by STXM-NEXAFS is sample S4-1. Also check the values that did not match the ones in Table S2.

Thanks for pointing this out. We have corrected the sentence and Table S2.

> RL355-356: "In the S3-3 and S4-2 samples, OCIN particles are dominant (~87.8 % and ~98.8 %, respectively), and there is only a very small fraction of OC-rich particles (~5.2 % and ~1.2 %, respectively)."

18. Line 344. "Figure 5 shows violin plots of the 'corrected' aspect ratio (left) and representative tilted images (right) for Pico 2014 (a to c), Pico 2015 (d to i), and Pico 2017 (j to q)." The description here is different from the caption of Fig. 5. Correct it.

We have corrected the caption of Fig. 5 as below:

> **"Figure 5. Violin plots of 'corrected' aspect ratio (left) and typical tilted images (right) of Pico 2014 (a to c), Pico 2015 (d to i), and Pico 2017 (j to q). Distributions in the left panels of (d to i) are the aspect ratio of Pico 2015 particles collected on stages 3 (left) and 4 (right). The shaded region corresponds to the different phase states (red: solid state; green: semisolid state; blue: liquid state). The red lines indicate the means, and the black dots the medians."**

19. Line 368. "The substantial fraction of solid and semisolid particles might be less oxidized" In Table 1, I found that SA1 and S6, whose average aging time is both longer than 16 days, have smaller fraction of liquid particles than other samples. Can you explain why the fraction of liquid particles is smaller with longer aging time?

We appreciate this great question from the reviewer. We wanted to provide a potential hypothesis for the phenomenon we observed, and future studies might need to confirm our hypothesis. We hypothesized that the substantial fraction of solid and semisolid particles might be less oxidized in the FT than the liquid particles during transport. We revised the statement as follows.

> RL404-404: "Thus, we hypothesize that a substantial fraction of solid and semisolid particles might be less oxidized and less prone to be removed via aqueous-phase processes than liquid particles in the FT during transport."

20. Line 379. Change "5(a, d, e, I, and j to o))" to "5(a, d, e, i, and j to o))"

The correction has been made as below:

> RL423-426: "In addition, we observed two different types of aspect ratio distributions: (a) narrow distribution with mean aspect ratios below 4 and a smaller fraction of particles (< 35 %) with aspect ratios greater than 4 (standard deviations of aspect ratio were ranging from 0.9 to 2.1) (Fig. 5a, 5d, 5e, and 5i to o), and (b) broad distribution with a larger fraction of particles (> 40 %) with aspect ratios greater than 4 (standard deviation of aspect ratio were ranging from 1.4 to 2.4) (Fig. 5b, 5c, 5f, 5g, 5h, 5p, and 5q)."

21. Line 383. "For S4-2, a possible reason is that the volatile and less viscous species of particles collected on the TEM grid have already evaporated and left these tiny residuals around those big particles (see Fig. 5(f) right panel) due to difference in temperature, RH, and pressure between OMP and SEM chamber." Does this problem also exist in the experiments of other samples?

    This is a great question. Based on the tilted images of all samples, we did not see other samples having the same issues since we did not observe tiny residuals around the particles. We added the discussion of limitations of the ESEM experiments in the revised manuscript RL168-174:

    > RL168-174: "Ambient particle samples were analyzed with ESEM at 293 K, under vacuum conditions (~2×10-6 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."

    > **Moreover, we also revised the sentence below to make this point clearer to readers:**

    > RL430-433: "For S4-2, a possible reason is that the volatile and less viscous species in particles collected on the TEM grid have already evaporated and left these tiny residuals around those big particles (see Fig. 5f right panel) due to difference in temperature, RH, and pressure between OMP and the SEM chamber, a phenomenon which has not been observed in other samples. Hence, those particles remaining on the substrate in S4-2 have a higher viscosity than the original particles."

22. Line 402. I did not see the viscosity of BBOA predicted in DeRieux et al. (2018) is up to 10^{12} Pa s. I suggest you only show what is the range of the viscosity under the atmospherically relevant RH. Add Li et al. (2020) who also calculated the viscosity of BBOA based on volalilaty distributions (Li, Day et al. 2020).

    We want to thank the reviewer for this comment. The RH values at OMP during our study range from ~6% to ~67% (see Fig. S2 and Table S1 and S2 in revision), we think it should still be valid to report these literature values since they are within the RH range at OMP during our study. Moreover, we added a sentence to discuss the viscosity of BBOA reported by Li et al., 2020:

> RL449-454: "DeRieux et al., 2018 predicted the viscosity of biomass burning particles using their chemical composition, and they found that the viscosity of biomass burning particles varied between 10^{-2} Pa·s and 10^{9} Pa·s depending on the RH. Liu et al., 2021 found that ambient and lab-generated biomass burning particles are in a non-solid state at RH between 20 % and 50 %. Li et al., 2020 predicted the viscosity of ambient biomass burning organic aerosols from volatility distribution and found that it varies between 10^{-2} Pa·s (liquid state) to above 10^{12} Pa·s (solid state) in Athens (Greece) and 10^{-2} Pa·s (liquid state) to ~ 10^{9} Pa·s in Mexico City (Mexico)."

23. Line 416. "Shaded areas represent regions of different phase states (liquid: blue, semisolid: green, and solid: red), with the boundaries of each region based on (O'Brien et al., 2014)." Can you give a more detaied explaination how to get the boundary lines?

    We appreciate the reviewer for this question. These boundary lines were based on previously reported field and lab-generated organic particles measurements described in O'Brien et al., 2014. To make this clear, we revised the sentence as below:

    > RL465-467: "Symbols are colored by their OVF. Shaded areas represent regions of different phase states (liquid: blue, semisolid: green, and solid: red), with the boundaries of each region based on measurements of field and lab-generated organic particles reported in O'Brien et al., 2014."

24. Line 430. "We used the density (ρ_{org}), hygroscopicity (κ_{org}), and dry glass transition temperature ($T_{g,org,0}$) of organic particles as reported by Schum et al., 2018 (see SI) since we do not have molecular compositions for our samples and Schum et al., 2018's samples were also collected at OMP during the same seasonal period (June and July).". The previous analysis in this manuscript mentioned that the composition of organic matter is quite different for different samples. Therefore, $T_{g,org,0}$ would be changed. There are three samples in the study of Schum et al. (2018), and the estimated Tg are also varied. Discussion of the uncertainties in Tg,org,0 is betted added.

    We thank you for this valid comment. Please see our response to major comment 1. We added the discussion of uncertainties in the revised manuscript.

25. Line 441, also cite (Schmedding, Rasool et al. 2020, Li, Carlton et al. 2021).

    We have added these two references. Thanks for suggesting them.

26. Line 490, cite (Li, Carlton et al. 2021, Shrivastava, Rasool et al. 2022).

    We have added these two references. Thanks for suggesting them.

27. Line 930. Change "solid black cycles" to "solid black circles"?

    We have revised Fig. 2 and its caption as below:

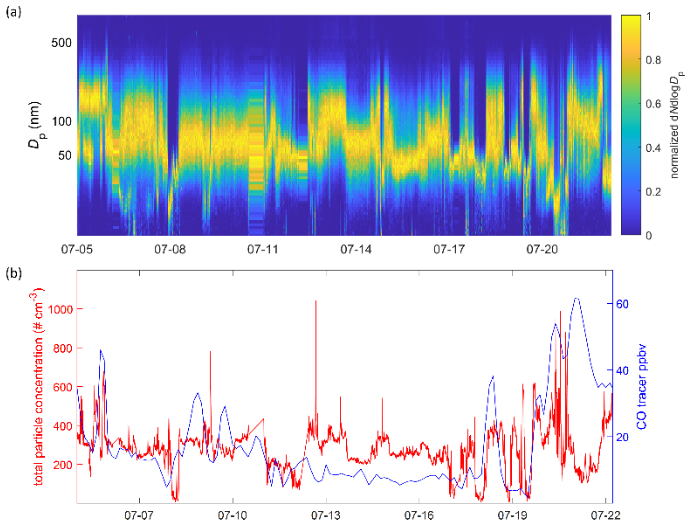


Figure 2. (a) Normalized particle size distribution from 10 to 800 nm measured from SMPS measurements, and (b) SMPS derived total particle concentrations (left y-axis, red line) and CO tracer retrieved from FLEXPART simulations (right y-axis, blue line) from 05 to 21 July 2017.

28. There is no need to use italics in the columns 12 and 13 in the first row in Table 1.

Thanks for this. We have corrected this issue.

29. What does the colorbar in Figure 1 indicate?

We added the flowing sentience to the caption of Fig. 1 and Fig. S7 in revision:

> **"The color bars indicate the ratio of column integrated residence time to the maximal residence time in the logarithmic scale. The X-axis and y-axis represent latitude and longitude, respectively."**

30. The inserted figures should be described in the caption of Figure 3.

Thanks for this comment. We revised the caption of Fig. 3 as below:

> **"Figure 3. Chemically-resolved size distributions were inferred from the CCSEM-EDX data for 2017. (a) Fraction of different particle types for all samples. Normalized chemically-resolved size distributions of (b) S3-1, (c) S3-2, (d) S3-3, (e) S3-4, (f) S4-1, (g) S4-2, (h) S4-3, and (i) S4-4. Inserts represent the normalized number fraction of different particle types as a function of particle size."**

31. Change "SA1" to "SA2" for panel b in Figure 5.

Thanks for pointing this out. We have corrected this in Fig. 5.

Supporting Information：

32. Line 2. *The title in the supplementary is different from the title in the manuscript.*

    We attempted to correct all the errors in the updated SI. We also corrected the title.

33. Line 21. “where Tg,w is equal to 136 K, is the Tg for pure water”. Cite (Kohl, Bachmann et al. 2005).

    We have corrected these two sentences as below:

    > RSL51-52: “where $T_{\mathrm{g,w}}$ is the T_{g} for pure water, k_{GT} is the Gordon-Taylor constant, κ_{org} is the CCN-derived hygroscopicity parameter of the organic fraction, ρ_{org} and ρ_{w} are the density of water and organic material, respectively.”
    >
    > RSL58-60: “Moreover, k_{GT}, $T_{\mathrm{g,w}}$, κ_{org}, and ρ_{org} were assumed to be 2.5 (Shiraiwa et al., 2017), 136 K (Kohl et al., 2005), 0.12 (Schum et al., 2018), and 1.4 g cm^{-3} (Schum et al., 2018), respectively.”

34. Line 29. “Moreover, k_{GT}, $T_{g,w}$, κ_{org}, and ρ_{org} were assumed to be 2.5 (Shiraiwa et al. 2017), 309 K (Schum et al. 2018), 0.12 (Schum et al. 2018), and 1.4 g cm-3 (Schum et al. 2018), respectively.” Why 309 K is for Tg,w? Check it.

    We have corrected this as below:

    > RSL58-60: “Moreover, k_{GT}, $T_{\mathrm{g,w}}$, κ_{org}, and ρ_{org} were assumed to be 2.5 (Shiraiwa et al., 2017), 136 K (Kohl et al., 2005), 0.12 (Schum et al., 2018), and 1.4 g cm^{-3} (Schum et al., 2018), respectively.”

35. Figure S2. What the x-axis stands for in figures b to r?

    The X-axis shows the particle number. To make this clear, we revised the caption of Fig. S2 (Fig. S3 in revision) as below:

    > **“Figure S3. Relative element percentage of 15 elements (C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Mn, Fe, Zn) for (a) average relative atomic ratios for all samples, (b) SA1, (c) SA2, (d) SA3, (e) S1, (f) S2, (g) S3, (h) S4, (i) S5, (j) S6, (k) S3-1, (l) S3-2, (m) S3-3, (n) S3-4, (o) S4-1, (p) S4-2, (q) S4-3, (r) S4-4. The X axis indicates the particle number.”**

36. In Figure S2 and Figure S3, are the relative atomic ratios of elements same as the relative element weight?

    We are sorry for the confusion. To make it clear, we changed that to element percentage and modified the entire manuscript to keep it consistent. Thus, the caption of Fig. S3 (Fig. S4 in revision) has been revised as:

> **"Figure S4. Flow chart to classify Pico 2017 particle types based on their element percentage retrieved from CCSEM/EDX measurements."**

37. Figure S4. Change "Jun" to "June".

    Thanks for pointing this out. We have revised that.

38. In Fig. S5-S6, I don't understand why the residence time is in percentage and how did you calculate it?

    In these results, residence time has been integrated vertically for the entire transport time (20 days) and the whole atmosphere. The residence time shown is color-coded in the logarithmic scale representing its ratio to the location of maximal integrated residence time (100 %). The reason for doing this is to simplify the comparison between two transport cases because the value of maximal integrated residence time for each transport case can be largely different. We can easily tell the relative time a plume spends over land vs. ocean and have a clear view of the transport pathway. To make this clear, we revised the caption of Fig. S5 and S6 (Fig. S6 and S7 in revision) as below:

    > **"Figure S6. The vertical distribution of the retroplumes residence time at given upwind times retrieved from FLEXPART retroplumes for (a) S3-1, (b) S3-2, (c) S3-3, (d) S3-4, (e) S4-1, (f) S4-2, (g) S4-3, and (h) S4-4 for Pico 2017. The color bar represents the ratio of residence time to the highest residence time across the height scale at each upwind time. The black lines indicate the average height of the plumes during transport."**
    >
    > **"Figure S7. Column-integrated residence time over the 20-day transport time and the vertical distribution of the retroplumes residence time at given upwind times retrieved from FLEXPART retroplumes for Pico 2015. (a, b) S1, (c, d) S2, (e, f) S3, (g, h) S4, (i, j) S5, (k, l) S6. For panels a, c, e, g, and i, the color bars indicate the ratio of column integrated residence time to the maximal residence time at each upwind time in the logarithmic scale, and the X-axis and y-axis represent latitude and longitude, respectively. For panels b, d, f, h, j, and l, the color bars represent the ratio of residence time to the highest residence time across the height scale at each upwind time, and the black lines indicate the average height of the plumes during transport."**

39. In the caption of Figure S10, "Mean ambient temperature (blue) and the predicted RHdependent Tg,org values (green)". The ambient T is actually in green and Tg,org is in blue in the figure.

Thanks for pointing this out. We have revised the caption of Fig. S11:

> **"Figure S11. Mean ambient temperature (green) and relative humidity (RH) (red) extracted from the GFS analysis along the FLEXPART modeled path weighted by the residence time and the predicted RH-dependent $T_{g,org}$ values (blue) for (a) S3-1, (b) S3-2, (c) S3-3, (d) S3-4, (e) S4-1, (f) S3-2, (g) S4-3, and (h) S4-4. The blue and red shaded areas represent one standard deviation of ambient temperature and RH from the GFS analysis. The green shaded areas represent uncertainties of predicted $T_{g,org}$ estimated from the range of $T_{g,org}$(RH = 0%) and uncertainties in RH."**

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