The revised manuscript „Molecular and physical characteristics of aerosol at a remote marine free troposphere site: Implications for atmospheric aging" by Schum et al. has improved compared to the initial submission. The authors immediately corrected the misassigned back-trajectory analysis in Fig.1. They evaluated the possible bias in calculating the glass transition temperature by small weight molecules, which are lost during sample preparation. The authors show that small weight molecules, such as oxalic acid, only marginally influence the overall glass transition temperature. However, other small organic acids (acetic acid, formic acid) do have a pronounced influence on the glass transition temperature.

One main point noticed in the earlier discussion was that the authors discuss the differences of DI-ESI-signatures of samples with different emission sources, and conclude that the conditions during transport to PMO are the cause of the different chemical signature. The authors have addressed this point by acknowledging that the emission sources on their own do play a role (l. 504). However, in my opinion, the authors still do not adequately discuss this aspect throughout their current manuscript (except the parenthesis in l. 504). What is still missing is a discussion on possible secondary gas phase processes in an anthropogenic pollution plume (sample PMO-2), in which high NOx, high SO2, and secondary ozone can also result in completely different conditions, compared to the biomass burning plumes (PMO-1 and 3). In their response to the reviewer´s comments, the authors argue that aqueous phase processing "leads to SOA production with a greater array of carbon numbers; the greater number of carbon numbers matches more closely with our observations of a continuum of carbon numbers from 2 to 33 in PMO-2". This argument does not seem convincingly to me, since there is a clear continuum of carbon numbers in the same range for PMO-1 and PMO-3 (Fig. 4 (a)-(c)), as well. The continuum of carbon numbers is actually the largest in PMO-1 (the biomass burning sample), resulting in ion signals up to 700 amu (Fig. 2 (a)). Thus, it looks like the emission source rather dominates the observed carbon number array.

I like the idea of comparing the PMO samples with samples of cloud and fog water (Table S.6) to identify compounds that are unique markers for aqueous phase processing. While the comparison between Cook et al. (2017) and PMO shows in fact the largest number of signals in common with PMO-2, the comparison with Zhao et al. (2015) shows the majority of common signals with PMO-1. We see that O/C of the comparison is highest for the sample PMO-2, but it remains unclear whether this is driven by the stronger presence of high O/C compounds in PMO2, which are not present in the samples PMO1/3. Thus, to me it remains elusive that this comparison is a clear indication for the "influence of aqueous phase processing" in sample PMO2.

Concerning ESI-artifacts: I do believe that negative ESI is less prone to adduct artifacts than positive ESI. The authors mentioned that samples are diluted to the lowest possible level to obtain a stable current during ESI. In principle, ESI should form always a stable current- even without any analyte present (e.g. in blank measurements). The authors mention that their AGC target of 1e6 ions was reached after 20-80 ms. I compared this to injection times on a Q-Exactive System (AGC target 1e6) after chromatographic separation – same ballpark. Thus, it seems that the samples were in fact reasonably diluted, however, it would also be interesting to see the actual ion count rates. I am asking for these numbers, since the NL value of the MS/MS experiments seem to me rather low (e.g. 5e3 for m/z 300 +/- 3). Such low value can be an indication for the presence of cluster ions, provided that the ion signal in the full scan shows a much higher signal (not possible to evaluate here, since the numbers are not reported).

Overall, I see the strength of this paper in relating the extracted conditions (T and RH) during transport with the observed chemical composition. But, I do miss a solid chain of reasoning toward the hypothesis of aqueous-phase aging in the MBL in contrast to slow aging rates in the FT when the aerosol is in a glassy state. It all goes back to the fact that the samples have a completely different source (anthropogenic pollution vs biomass burning), and the chemical signature of the aerosol, when it is close to its emission source (t_zero of aging), is not known. Thus, I do not see point 5 of the ACP review criteria fulfilled (*Are the results sufficient to support the interpretations and conclusions?*). Still, the paper provides a novel and interesting concept: the extraction of glass transition temperatures from molecular ion measurements. Also the atmospheric implications are highly relevant: slow aging rates of BB aerosol and slow decomposition of BrC in the free troposphere. Therefore, I recommend the article to be published in ACP after a more critical discussion regarding the fact that the initial aerosol composition in plume PMO2 (when it is still over the continent) is not known, and that other processes in the plume than only aqueous-phase oxidation during transport could explain the high O/C of the sample PMO-2.

Specific comments:

l. 13-16: Here the authors conclude that "environmental factors during transport" are responsible for the higher O/C in PMO-2. I am missing any mentioning of the possibility of different chemical composition (e.g. already higher O/C) of the PMO2 plume right at its source. Also, the term "environmental factors during transport" is too blurry.

l. 20: The same issue here: the comparison between PMO1/3 and PMO2 has to be discussed with more caution. The data presented do not allow the conclusion that solely Tg/T is the cause of higher oxidized aerosol in PMO2. I recommend something like "[…] and therefore less susceptible to oxidative aging than the organic aerosol transported in the boundary layer."

l. 28: […] cloud droplet and ice nucleation activity […]?

l. 34-36: The biomass burning studies cited here might not be the best, since they all refer to biomass burning events from grassland fires. Grass-lignin is different from softwood (coniferous) lignin, resulting in different biomass burning marker molecules (Simoneit et al., 1993). Refering to studies on biomass burning plumes from boreal forest fires (e.g. Corrigan et al. (2013)) might be more appropriate in the context of discussing plumes from Canadian boreal forests. It is well recognized that the most prominent organic biomass burning marker (levoglucosan) undergoes fast photooxidation, however, I did not find a connection between biomass burning markers and their oxidative degradation in the paper by Vakkari et al., 2014. Here, a paper on the degradation kinetics of levoglucosan would fit much better (e.g. Lai et al. (2014), Arangio et al. (2015))

l. 38: The authors might find our paper on long-range-(low-altitude)-transported biomass burning aerosol from wildfires in Russia to the SMEAR station in Hyytiälä, Finland of interest. During a pollution plume event we observed highly oxidized aerosol (O/C ~ 0.70), while the average O/C during this campaign ranged around 0.5. We also observed high-molecular weight organic matter in the aerosol phase during this BB event, indicated by molecular ion signals up to *m/z* 800 (Vogel et al., 2013).

l. 244: A low oxalate/sulfate ratio is reported for PMO-2, likely obscured by high sulfate concentrations. The authors argue that oxalate is also high in PMO-2 due to aqueous phase processing. To further search for evidence that cloud processing has really occurred for PMO-2, it would be of interest to compare the oxalate/sulfate ratio in sample PMO2 with reported oxalate/sulfate ratios in the Eastern US close to the emission sources.

l. 307: Is the North American SOA solely anthropogenic? I think there are several studies reporting dominant biogenic SOA in East America (e.g. isoprene SOA in South East US?).

l. 353-366: The authors describe a higher O/C in PMO2 for the CHOS group compared to PMO1, and argue at the end of the section that this observation highlights the enhanced aging during transport of PMO2. While I believe that there is indeed a higher oxygen content in the sample PMO2, it does matter to which atom the oxygen is bonded. If it appears as oxygen-sulfur bonds (as organosulfates), then the increased oxygen content (higher O/C) in PMO-2 rather tells us that the different source emissions of the PMO2 plume allowed enhanced formation of organosulfates. This observation would be in line with the observation of higher inorganic sulfate in PMO2, as well as the expectation that SO2 emissions over North East US are higher than in the remote Canadian boreal forests.

l. 380: It is too speculative to talk about the CCN ability of the sampled aerosol particles when their size distribution is not known. Also the speculation about the amount of less volatile components seems to me ambiguous, since the total mass concentration available for gas-to-particle partitioning will also affect the fraction of higher-volatility compounds in the particle phase.

l. 472: What is about the role of multiphase and heterogeneous oxidation of the aerosol additionally to aqueous-phase processing of cloud droplets?

l. 480-494: The section is missing a more critical discussion, including the fact that the initial chemical composition of the plume sampled in PMO2 is not known. Again, PMO2 is compared here against PMO1 and 3. The fact that higher sulfate is observed in PMO2 compared with PMO1/3 rather goes back to the emission source, where you expect more SO2 being emitted in North America than in the boreal forest. The relatively higher abundance of sulfate in PMO2 against PMO1/3 hence <u>does not</u> necessarily support cloud processing.

l. 488-491: These lines are redundant with section 3.1 (l.235 ff.).

l. 508: Only aqueous-phase oxidation?

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