

Dear Authors:

Thank you for your response to the referee comments and your manuscript revisions. Both referees expressed that the manuscript would be strengthened by further calculations/analysis and one felt that it should be recharacterized as a measurement report if no further analysis was performed. Given the unique aspects of the dataset and the analysis included, I support publication as a research article. After carefully considering all the documents, it is my opinion that several revisions are required before the manuscript is suitable for publication in ACP. Several of these relate to instances where responses to referee comments were informative in the response document but were not included in edits to the main document. Please consider the comments below. Unless otherwise noted, line numbers refer to the track changes version of the manuscript.

Dear editor,

Thank you for your helpful comments. We have addressed them below in grey italics.

Main comments

1. I suggest you consider rewriting many of the chemical formulas to better reflect the ions. For some (non-exhaustive) examples H_2O_6^- is probably better written as $(\text{HIO}_3)\text{IO}_3^-$, $(\text{SO}_4)\text{H}_3\text{O}^-$ is probably better as $(\text{H}_2\text{O})\text{HSO}_4^-$, $(\text{SO}_4)_3\text{H}_5^-$ is better as $(\text{H}_2\text{SO}_4)_2\text{HSO}_4^-$, etc. This will be more aligned with how these ions have been represented in past works and also increases the ease of reading.

We have gone through the document and tried to improve this.

2. Section 2.1: Please add more details regarding the PSM even though more extensive details are given in the cited work. It is important that the basics are conveyed without having to reference another manuscript. Something along the response given to Dr. Brean's comment regarding line 98 (of the original document) is sufficient.

We added the text: "The PSM was first used in fixed mode with a saturation flow rate of 1 lpm and from 17 September it was used in fixing mode, with the saturation flow switching from 0.1 to 1 lpm every 60 s. Saturation flow rate of 1 lpm was assumed to correspond to approximately a cutoff diameter 1 nm and 0.1 lpm to 3 nm."

3. Line 117: Please add a sentence or two similar to the response given to Dr. Brean's comment regarding line 117 (of the original document) regarding the similarities/differences to PSCF.

We added the text: "This method is similar to the Potential Source Contribution Function (PSCF) method described by Fleming et al. (2012), but more simplified. One of the major differences is that we do not use a specific threshold for high concentrations like the PSCF but use an average of all concentrations."

4. Line 144-145: "... $(\text{NH}_3(\text{H}_2\text{SO}_4)_3\text{HSO}_4^-$, in the peaklist unknown0085)." is confusing wording to the reader. I suggest that you make it clear that this peak was assigned later on and provide the ppm difference between the assignment and the unknown. Please also include the assignment in the peak list.

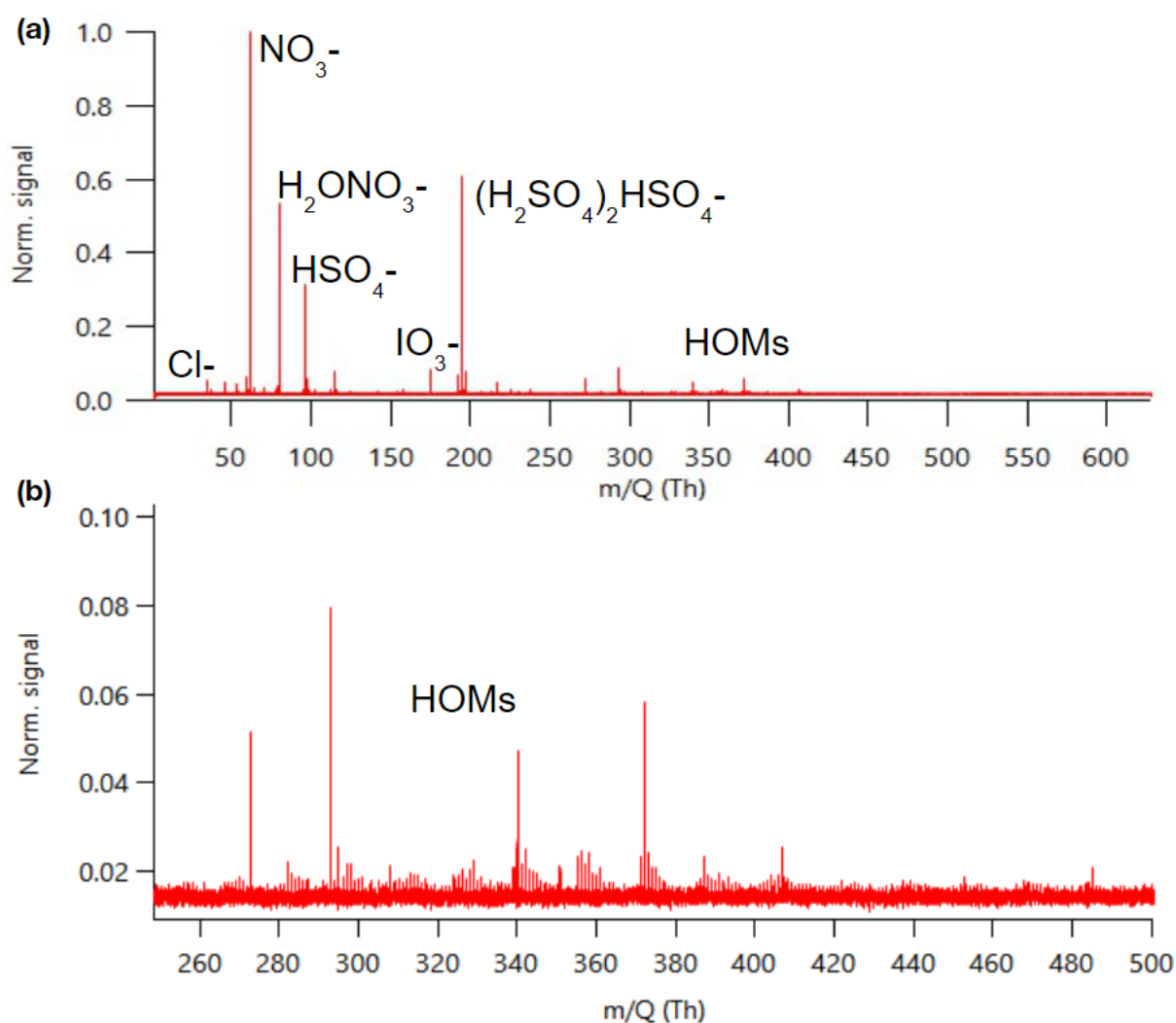
We have replaced the text “in the peaklist unknown0085” by “please note that this peak was only identified after processing the data and that is why the mass of the peak is 13 ppm off” and changed the name in the peaklist from unknown0085 to the expected composition.

5. Lines 153-155: The wording here makes it sound like the lack of peaks can be quantitatively explained by these factors. I suggest rewording to make it clear these are possible reasons.

We replaced the text “The lack of peaks at higher masses can be explained by a combination of” by “Possible reasons for the lack of peaks at higher masses include”.

6. Figure 1: I suggest considering enhancing the intensity of the peaks above approximately m/z 250 by using a scaling factor. Alternatively, a log scale y-axis may make it easier to see some of the peaks.

We thought about this and ended up adding a second panel that shows the masses between 250-500 m/Q better (see below) Masses above 500 m/Q were not included, since there were no peaks and this way the peaks in this range are more clear.)



7. Figures 2 & 3: Personally, I found the examples presented in the response to questions posed by both referees more compelling as it further enhanced the strong relationships that are discussed in the manuscript. Particularly the strong positive

8. Line 295: Following up on Dr. Brean's comment, the diel profile may be more consistent with organonitrate production from NO₃ radical chemistry than from OH + NO_x chemistry. This could plausibly explain the difference in the diel profile between your work and the Bianchi et al work.

Yes, this sounds possible. We added text “, potentially organitrates produced through NO₃ radical chemistry and clustered with a nitrate anion (see e.g., Yan et al, 2016)”, to the end of the paragraph.

9. Line 355: I suggest including the further information on the attribution of non-sea-salt sulfate from DMS and SO₂ from Australia provided in response to Dr. Jokinen's comment on line 344.

We added text “The previous work by Li et al. (2018) estimated that DMS contributed to 73–79% and SO₂ emissions from shipping activities ~21–27% of the non-sea-salt sulfate when taking into account data from all source area sectors. We assume that the ratio would be similar for the bisulfate dimer.”

10. Figure 8: I don't understand why from ~12:00-19:00, the N1-10 timeseries in the lower panel of (a) is at zero while the upper panel shows high counts in the 2-10 nm region for most of the time period and N1-3 is also non-zero. Is it because these are all from different instruments? If so, how does this disagreement impact conclusions/interpretations about particle counts at these lowest sizes throughout the manuscript?

Yes, as stated in the caption, these are all from different instruments that have different inlets and function differently. The N2-10 range of the upper panel is using NAIS data whereas N1-10 is calculated using both PSM and SMPS data and N1-3 using only PSM data. Both PSM and SMPS count particles with CPC's that grow the particles by condensing butanol on them and then count them optically. The NAIS on the other hand uses unipolar chargers, differential mobility analyzers and electrometers to determine the particle size distribution. In the NAIS, the losses of small particles are clearly lower since the NAIS has a higher flow rate (60 lpm) and larger inlet (diameter 2.5 cm) tubing whereas the inlets for PSM and SMPS were only ¼" wide and had an order of magnitude lower flow rates.

Since N1-10 uses two different instruments which can have different sensibilities it is more uncertain and it is possible for the value to go to zero. On the other hand N1-3 also has its own uncertainties since this is a rather narrow size range and the PSM is sensitive to environmental conditions and the losses of these small particles are higher than those of larger particles.

In Section 3.5 where we connect aerosol data to APi-TOF data we focus on SMPS data and 2-4 nm anions from NAIS data for land-influenced NPF and PSM and SMPS data for marine NPF and we do not really use the neutral NAIS data to interpret the APi-TOF data.

In the beginning of Section 3.5.2 we mention ‘Our previous work showed that in clean marine air, new particle formation should not be studied with the traditional criteria used for continental sites. Instead we focus on particle growth episodes and appearance of sub-10 nm particles. -- We also show PSM data for sub-10 nm particles instead of NAIS data, since NAIS seemed to underestimate particle concentrations which is especially problematic in the clean marine air where the concentrations are low.’ We have now

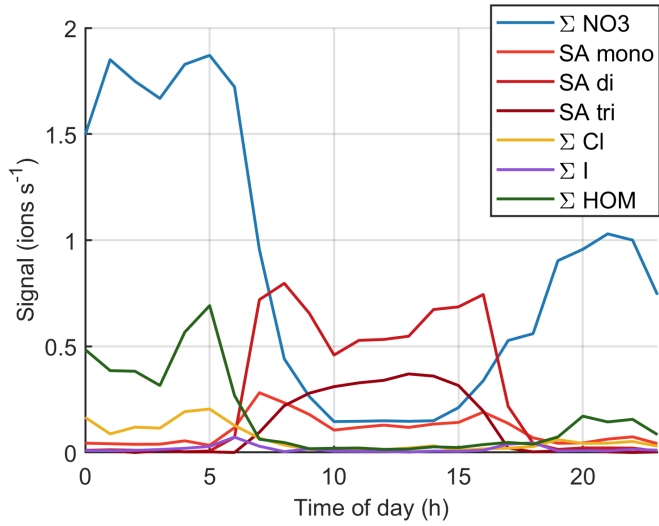
added text ' Here it should be noted that the NAIS and PSM have different functioning principles and hence also different uncertainties. In the PSM data,.' after this to alert the reader of the uncertainties in the data. Throughout the text there are also several mentions of when the 2-4 nm anions and N1-10 are in agreement and when not and we hope that the reader can make their own conclusions on this.

11. Discussion starting at line 423: API-TOF is best suited for capturing NPF happening at the measurement location and thus events identified as transported events in your previous work would likely not be observable as the characteristic NPF API-TOF peaks. I think it would strengthen the manuscript to include some of the information provided in your previous manuscript about if any/all of these were classified as transported events by your previous analysis and thus might explain the lack of larger ions observed by the API-TOF. Are there differences in the ions observed between transported and regional event days?

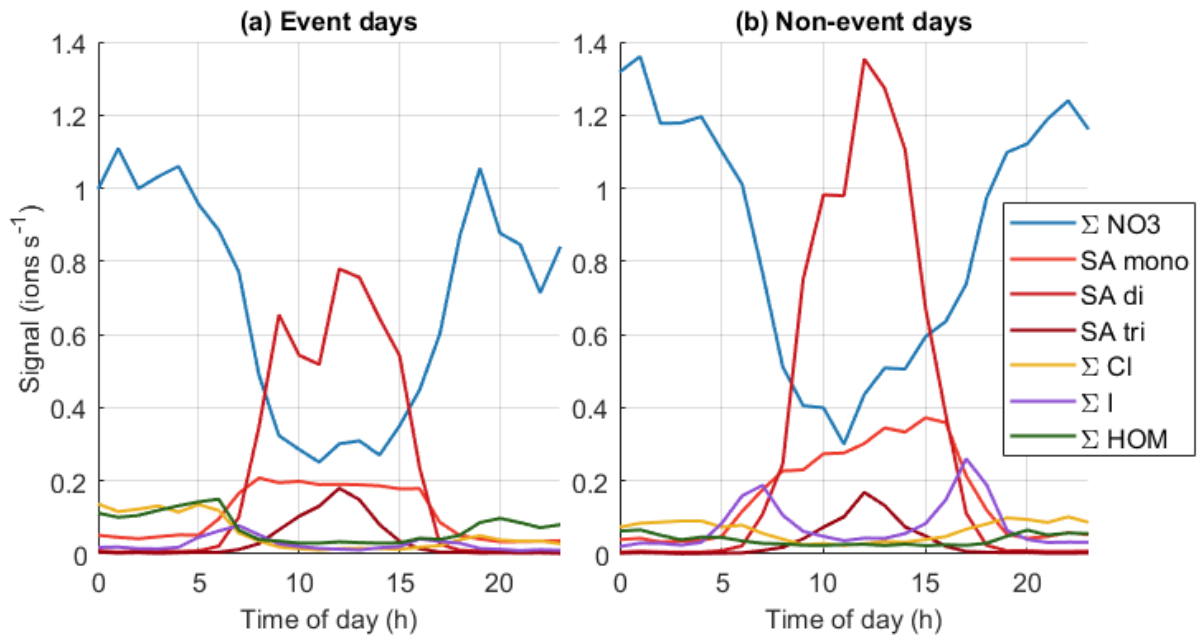
This is an interesting point. The figure that compares event and non-event days considers event days to be Class I and Class II events. Our previous work showed that out of Class I 30% were transported events whereas out of Class II events only 10.5% were classified as transported events (when data from both classifications was available). In total that makes 20.5% of the events considered here, so presumably the figure should be dominated by regional events. We did a quick check to see if the levels of different compounds (here just HSO₄⁻, CH₃O₃S⁻, IO₃⁻, C₁₀H₁₄O₉NO₃⁻) differ on regional and transported event days, but did not see any statistically significant differences, so we expect that the transported events do not significantly change the results.

12. Figures 9 and 10: I agree with both referees that it would be nice to see this version in a non-stacked view so that the reader can more clearly see the diel variations. I agree with your point in the response that the figure is too busy when all of the ions are included. However, many of the ions/ion groups are extremely low and poorly resolved in the current figure as well. I think a simplified figure including the most important ions (either because of overall intensity or because they are discussed substantially in the manuscript) would be appropriate. Placing such a figure in supporting information would be appropriate. In Figure 10, please state the number of event and non-event days.

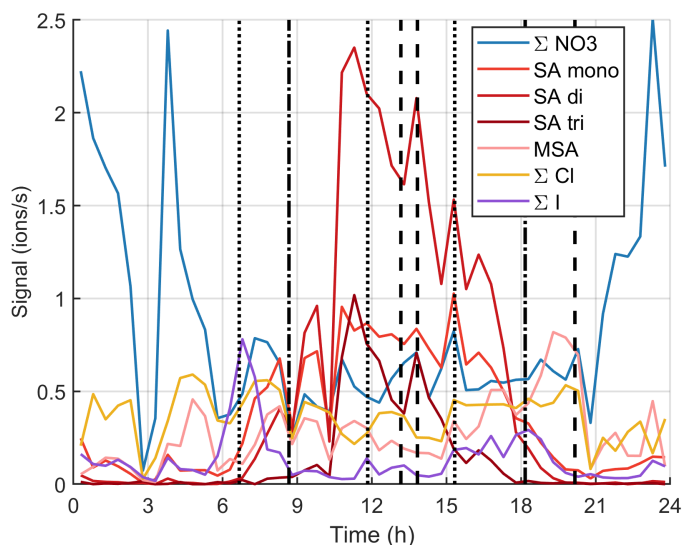
We have made simplified plots for Figures 9 and 10 and for consistency also figures 12 and 13 and added them to the supplement (see below). There were 53 event days and 90 non-event days with APi-TOF data available and we have added this information to the caption of Figure 10.



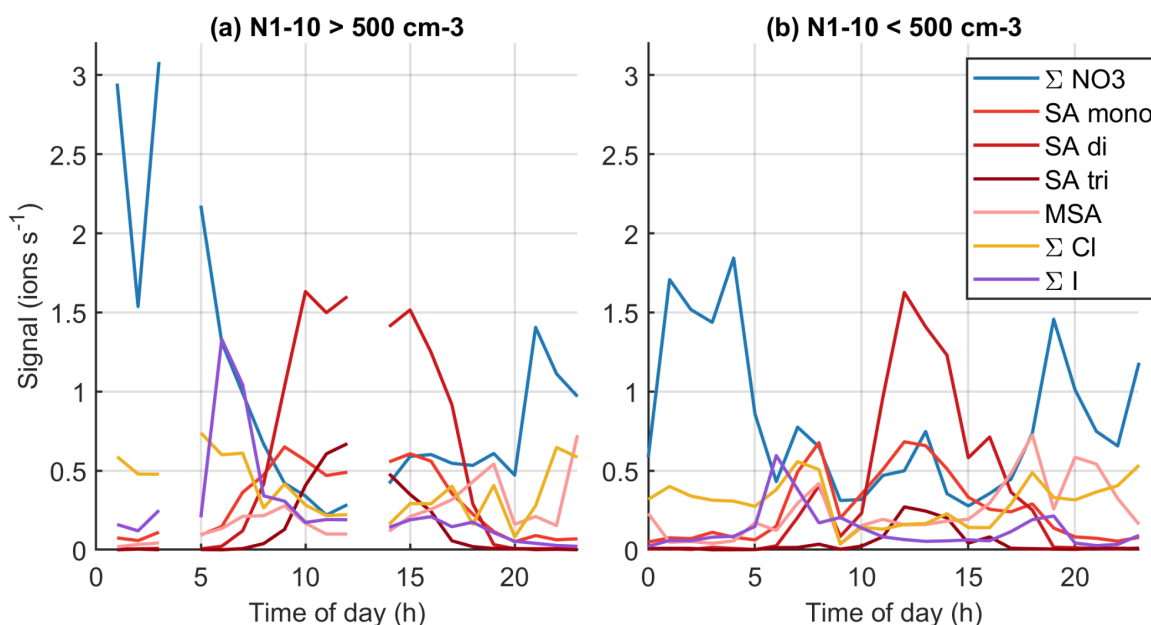
Additional figure for Figure 9.



Additional figure for Figure 10.



Additional figure for Figure 12.



Additional figure for Figure 13.

Technical

1. Line 106: Please specify the inner and outer diameters of the tubing. Please also provide the flow rate into the instrument.

We have replaced “3/8” thick stainless steel inlet” by “stainless steel inlet with an outer diameter of 3/8” and thickness of approximately 1 mm” and added text “The instrument flow rate was 0.8 lpm, but” to the beginning of the next sentence.

2. Both m/Z and m/Q are used in the manuscript. Please standardize.

Thank you for pointing this out. We have changed all m/Z to m/Q.

3. Lines 169-170: Everything in the list with the exception of HOMs are given in their ion form. I suggest saying identifying HOMs as the observed ion (e.g., HOMs clustered with NO₃-).

Done.

4. Lines 257-258: Please include units on the condensation sink values.

Done.

5. Figure 6: Please include a legend for land and marine. I also find this figure very difficult to read as the panels are small and the seasonal variations are muted due to the use of a log scale. In figure 5, the diel cycles were more apparent making the figure easier to interpret than this one. I suggest considering only showing the most important panels in Figure 6 in the main text and making them larger. The other ones can be put into supporting information. Would a non log plot help make the seasonal cycles more apparent?

We have added the legend, divided the figure into two figures and replaced text 'We use only 10 groups' by 'Here we show six of the most important compounds (Fig. 6) and in the supplementary four more compounds (Fig. B2).'

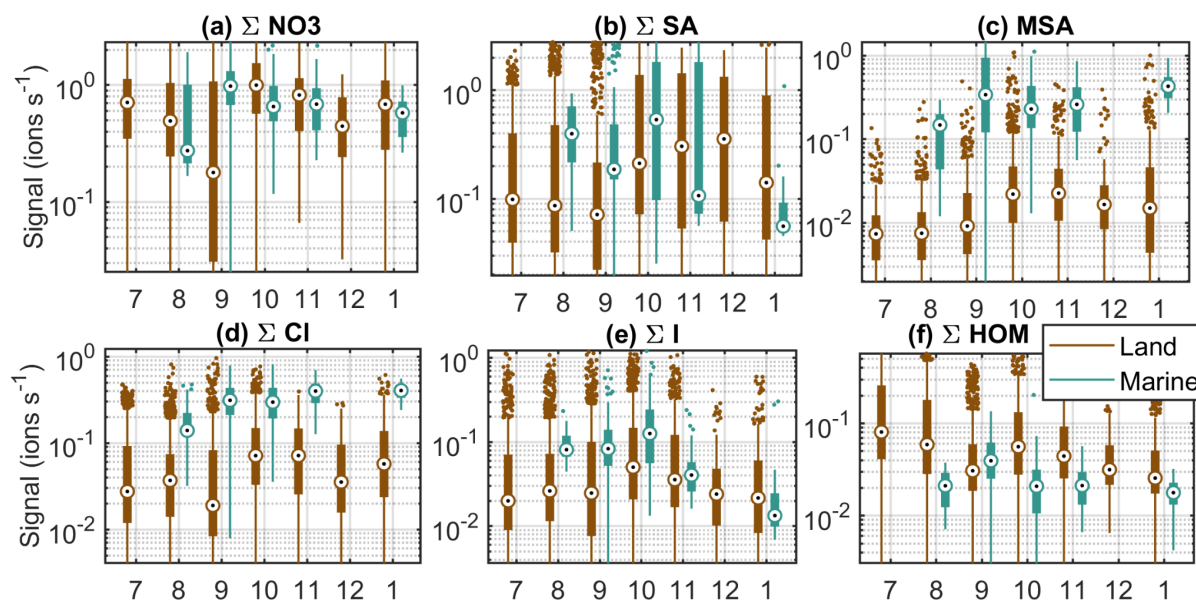


Figure to replace Figure 6.

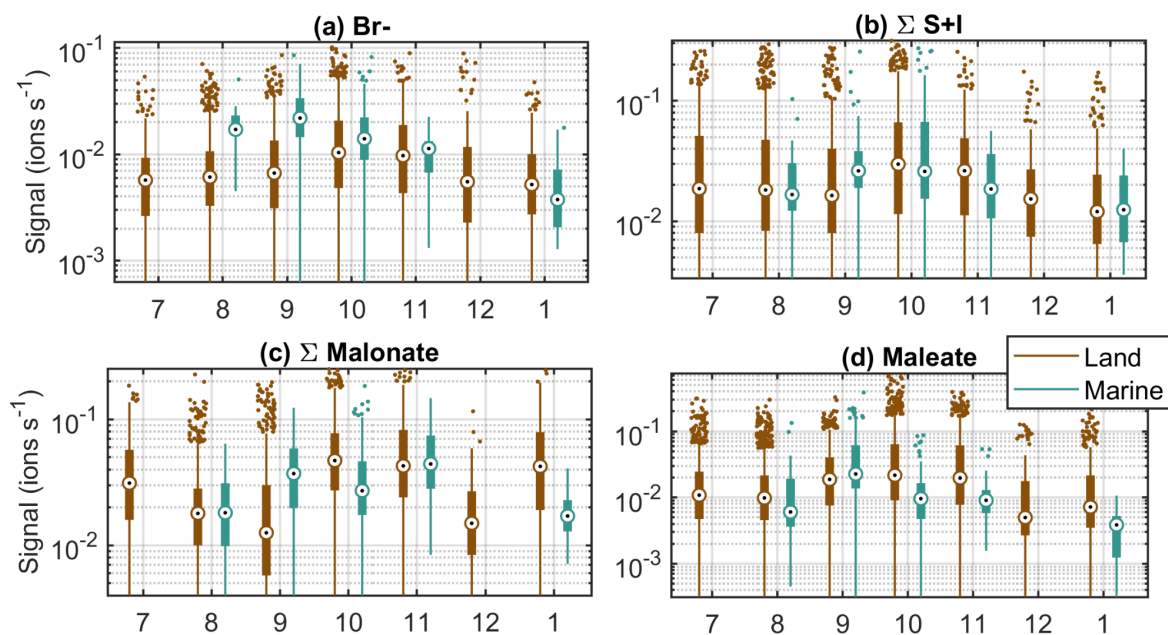


Figure B2 that will complement Figure 6.

6. Section 3.5.2: I think the name of this section should be modified to reflect that it discusses sub 10 nm particle days and not specifically true NPF.

In addition to sub-10 nm particles, this section also discusses later stage growth and even if we do not see traditional banana type events, which only occur in homogeneous air masses, we consider observing sub-10 nm particles as NPF.

References:

Yan, C., Nie, W., Äijälä, M., Rissanen, M. P., Canagaratna, M. R., Massoli, P., Junninen, H., Jokinen, T., Sarnela, N., Häme, S. A., et al.: Source characterization of highly oxidized multifunctional compounds in a boreal forest environment using positive matrix factorization, *Atmospheric Chemistry and Physics*, 16, 12 715–12 731, 2016.