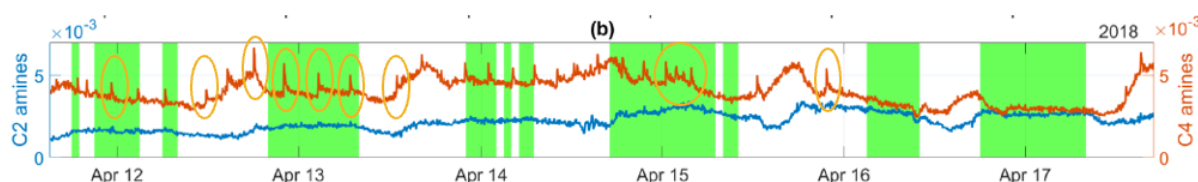


I sincerely appreciate that the authors have carefully considered the comments and concerns I had raised previously. Most of my comments have been addressed satisfactorily. I will elaborate on the two exceptions (Comments 1b and 3) below.

Otherwise, I have found the revised paper well structured and written, and following comprehensive and scientifically sound procedures for analyzing a novel ambient dataset. I believe re-addressing my remaining concerns will amount to only minor additional revisions, subject to which I gladly recommend the manuscript for publication.

Regarding "Comment 1b":

It may be worth clarifying unambiguously, which "spikes" I have been writing about: the spikes in the C4 amines time series, some of which I marked by orange circles I added below to the manuscript's Fig. 8b:



These spikes occur throughout the measurement period, both well within the white and green patches, representing BL and FT conditions respectively. Assuming we have all been talking about the same thing, the authors' response (and the manuscript at now P24 L15-16) contribute these spikes ("peaks") to mixing of BL and FT air "at the interface between the BL and the FT". My issues with that are:

- How to conclude that FT/BL mixing is happening in those instances? I would expect that to happen at the transitions between green and white patches, but the spikes occur throughout.
- If mixing was the cause of these spikes, what would be candidate mechanism? It would unlikely be a concentration differential between the different air masses, because that differential should at least to some degree become apparent once the BL grows to station elevation (or collapses beneath it). Could some evaporative process be responsible?
- Interestingly, as pointed out in the authors' response to "Comment 2", several other mass spectral peaks exhibit concurrent spikes (Fig. S14). Maybe I get too excited about those spikes now, but it is really unfortunate that those other peaks have remained unidentified, as their underlying compositions may reveal an explanation for what was going on.

With that in mind, I would like to ask again if the mass defect of those compounds could at least provide some clues regarding what kind of compositions are possible or can be excluded? They are mostly major peaks in the spectra. If m/z 198 and 261, for instance, are identifiable as $C_4H_{11}N.(HNO_3)_{1-2}.NO_3^-$, the mass defects of m/z 246, 260, etc. should at least be constrainable to a useful degree?

Alternatively, how certain are actually identifications like those for m/z 198 and 261?

So, to sum up my concerns here:

- 1) The connection of those spikes to BL/FT mixing, as currently put forward, requires further explanation.
- 2) If maintained, one or more mechanisms should be speculated on.
- 3) The involved mass defects (and/or uncertainties in peak identifications) should be at least discussed at some level.

Regarding "Comment 3":

First, I apologize that my sentence containing "ion clusters" was badly phrased (at the least I shouldn't have used the "ion clusters"). It was not meant as a separate comment, but only to explain why I believe the use of "cluster" is questionable in some of this paper's context -- as the authors anyway correctly understood based on the first part of their response. Indeed, I have no issue with the original axis labeling in Fig. 7b.

As for the actual point, the authors argue in their response that molecules identified by CIMS are commonly referred to as "molecular clusters". Could the authors give evidence of that "accepted usage in the literature"? (I have honestly failed to notice, and a quick Google search did not help either. Indeed, Google only serves me counter-examples. But even if evidence was provided, I would probably still argue that we should strive for using accurate terminology.) Anyway, I ended up believing that we are still/again misunderstanding each other here.

Better to continue with the added text that now points out the usage of the "cluster" term in Section 4.2.1.

That is a good addition, but it does not settle the issue. The added text (correctly) defines the "cluster" term as referring here to clusters present in the atmosphere and clusters formed in the instrument. That is of course correct and perfectly fine, but in contradiction to the authors' response and different from the (incorrect) usage of "molecular clusters" in the title and abstract (L24). Also incorrect, I believe in at P4 L7 (Intro), P34 L11 (Conclusions), and Table 1, which hopefully covers it all.

To reiterate, I still find the use of "clusters" in the title ("[...] molecular clusters in the free troposphere [...]") inappropriate, as it is about >90% erroneous (Fig. 7b). And it also misleads in the abstract, especially given the beginning with reference to new particle formation, which involves actual molecular clusters. I further suggest fixing those couple of other instances too. (Unless, that is, I have really been missing key literature that uses the cluster terminology as referring, confusingly, to molecules.)

And I appreciate the authors pointing out more examples of actual atmospheric cluster observations. So, for the title, for instance, just adding "and potential precursor molecules" could be an acceptable solution.