Interactive comment on “On-line determination of the chemical composition of single activated cloud condensation nuclei – a first investigation of single urban CCN and CCN obtained from sea water samples” by Carmen Dameto de España et al.

Anonymous Referee #1

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General comments:

The manuscript intended to introduce a novel method to characterize the individual activated CCN chemically. Based on a Cloud Condensation Nuclei-Versatile Aerosol Concentration Enrichment System (or CCN-VACES), particles can be firstly activated, followed by the detection. A Laser Ablation Aerosol Particle Time of Flight mass spectrometer (LAAPTOF) was deployed downstream to obtain the chemical composition of
individual CCN particles.

The experiment design and data analysis presented are quite well. However, the way that the authors presented their results might require improvement. The discussion on the results is full of details (several mass spectra of the detected particles) rather than presenting new knowledge out of such information. The authors need to provide an in-depth scientific interpretation and discussion on what is unique with this newly developed technique. Another major concern is that the major conclusions might not be supportive in the current version. More comparisons between individual activated CCN and individual droplet residues should be done to validate their results on the measurements of individual activated CCN. Barely with the mass spectral data, it is hard to confirm that the detection particles are in the form of droplets. Is it possible that the detected particles are already dried?

Overall, the topic of this manuscript is relevant to the journal and has importance scientifically. Prior to publication, the authors should also address the specific comments below.

Specific comments:

1. Introduction: the introduction of the CCN should be more specific for SSA particles. The authors just put some basic knowledge together, which is not explicitly in line with the major conclusion of this study. Some sentences, such as “Ambient aerosols originate from multiple different sources. Chemical reactions of natural and/or anthropogenic precursor gases lead to particle nucleation events.”, and “Water soluble organic carbon (WSOC) has been shown to influence particle activation (e.g. McFiggans et al., 2006, Jacobson et al., 2000).”, and “Cziczo et al. (2003) and Cziczo et al. (2006) coupled a continuous flow ice nuclei counter (Rogers et al., 2001) to a single particle mass spectrometer (PALMS; Murphy et al, 1998) to focus on chemical characterization of IN, and single particle analyses of ice particle residuals where conducted by Schmidt et al (2017) at Jungfraujoch.” are not necessary or duplicate.
2. Introduction: the authors mentioned that there are challenges to measure the chemical compositions of a single ambient particle. This might be not accurate. The development of SPMS could be dated back to decades, and there are many results in this topic, as also listed in the manuscript. Besides, Aerodyne aerosol mass spectrometer (AMS) could not address this issue.

3. Introduction: while there is no study to-date focusing on the chemical composition of single activated CCN, there are probably many results on the chemical composition of cloud or ice particle residues. Rather than listing the references, I suggest that the results related to chemistry compositions of SSA should be included to make the introduction more readable. Further, an answer to why direct measurements of droplets are essential is also necessary.

4. Introduction: as noted in Line 59, “The role of the contribution of organic material to SSA in remote regions”, more results on the observed of the chemical composition of single SSA in the atmosphere or cloud should be included to make it more complete.

5. Line 94 “…only a short time”: please be specific.

6. Instrumentation: It is not clear enough in the text to show how to separate the particles and droplets. Even if the cut size of the virtual impactor is 1.5 micrometer, the number fraction of particles with sizes larger than this should be estimated and accounted for in such measurements. In addition, the sizes of the produced SSA should be given to evaluate the property of droplet separation.

7. Again, how to test if the separated droplets are evaporated in the vacuum before being ionized?

8. Line 275: In the negative ion spectrum signs of O \((m/z=16)\) and OH\(^-\) \((m/z=17)\) correspond to water. References would be helpful here. Why were these peaks not shown in every droplet?

9. Figure 5-7: I do not understand why these peaks are present in the negative ions
spectrum.

10. Line 369: “The presence of Si+ (m/z = 28) in the spectrum in Fig.12 might indicate a CCN not originating in the area of Vienna.”. Such a statement is not satisfying and does not help in the discussion.

11. Section 5.2.2: Is there only one type of SSA? This is apparently different from previous laboratory studies.

12. Section 5.2.3: I would recommend the authors focus more on what is new about this mass spectra. Does it provide more information than those for droplet residues? Only with more validation, can the author state that such a technique would provide insights into the composition of individual activated CCN.