In their manuscript, "Chemical Composition-Dependent Hygroscopic Behavior of Individual Ambient Aerosol Particles Collected at a Coastal Site" the authors describe the deliquescence and efflorescence behaviour of ambient aerosol particles and correlate this with chemical composition. They also compare to a laboratory proxy system of known chemical composition and predict the deliquescence behaviour with AIOMFAC calculations. Using all of this data, the authors generate phase diagrams for sea spray aerosol, having complex chemical composition, through the deliquescence and efflorescence processes. I believe this work provides novel insight to the deliquescence and efflorescence behaviour of complex mixtures representative of ambient aerosol. Below I have detailed a few points that should be addressed and a number of comments to help with the readability of the manuscript.

Major Points

1. There are a couple key, but specialised terms in this manuscript that have not been defined clearly, making it difficult for a non expert to follow. I suggest explicitly defining eutonic composition and mutual DRH/ERH.

2. The manuscript is missing a description of AIOMFAC calculations. How did you go from model output to DRH? Were these calculations done for the lab generated aerosol only or also every unique composition of sea spray?

3. Could the authors please add some details about how the ambient aerosol samples were stored before analysis. Is there potential for changes in their chemical composition or morphology between collection and measurement?

4. Could the authors please comment on the use of nitrate salts but not sulfate salts in their sea spray mimic - in the introduction sulfate was listed as a component of SSA but nitrate was not.

5. Could the authors comment on their choice to use the ratio of ions in sea water. Divalent cations are known to be enriched in in sea spray aerosol (Jayarathne et al. (2016). Enrichment of Saccharides and Divalent Cations in Sea Spray Aerosol During Two Phytoplankton Blooms. Environ. Sci. Technol., 50(11511–11520). https://doi.org/10.1021/acs.est.6b02988)

Minor Points

1. Line 115 - 119 - This statement is not clear to me. It probably needs to be broken into 2-3 sentences. Is this the case for all binary salt mixtures or just some?

2. Line 242 - what are these exception particles chemical compositions?

3. Line 255 - are there figures for these particles?

4. Does see-through mean transparent? Consider making this substitution in the manuscript.

5. Section 3.2 seems redundant with the methods and the following sections. What is the take away message from this section that is not provided in the later discussion of the specific particle types?

6. I think for clarity, it would be helpful to give the lab generated SSA (surrogate system) a specific name and use it throughout. It seems like sometimes they are called surrogate systems and sometimes just systems, and it becomes hard to follow if it is the lab proxy or some ambient aerosol that has that composition.

7. Section 3.3 could use an intro sentence that reminds the reader that SSA is a fraction of the ambient aerosol.

8. Line 301 - 304 & 305 - 309 & 365 - 368 was there evidence of organic in the elemental analysis as well?

9. Figure 3 scale bar for optical images? It is hard to tell what is 20 μm in the final panel. The whole field of view?

10. Fig 4/5 - are the lines just joining the points or do they represent something?

11. There are many references to the AIOMFAC calculated DRH, but they are not shown anywhere. Consider tabulating these for the SI.

12. The captions for Figures 6 and 7 need to be modified so that the reader can understand what is going on in the figure without having to go back through the main text. For example, the two vertical lines are not defined and the phases are not easy to understand without the definitions for P, S, R, Q & T.

13. Could the authors please check that all the citations made in the text have an entry in the reference section. I noticed Liu et al. 2011 is missing but did not do a thorough check.