

The authors' have made a number of revisions, which have improved the paper, but have not fully addressed a number of issues raised by multiple reviewers. Due to the lengthy (and quite combative) nature of the response, only the most concerning remaining issues with the manuscript are addressed.

- **Limit of Detection:** With respect to using measurements below the limit of detection (LOD) of the system. The authors' have made the statement that "The analysis is consistent for Na⁺ concentrations over the full Na⁺ measurement range, including below our assigned Na⁺ LOD." If a measurement is below its LOD it is not considered reliable, since it is within a signal-to-noise ratio of 3 to 1. When the signal is near the intensity of the noise a number of factors can lead to issues. The authors' claim that researchers define LOD in different ways, but for a standard analytical measurement, such as chromatography, there is an accepted definition that is broadly used and which should be here. The fact that the data is consistent with data above the LOD, is still not a good reason for using this data. Frankly data below the limit of quantification (LOQ) (10:1) shouldn't be used, but signal should at least be > 3:1 to be used. The authors' do not seem willing to only use properly quality controlled data, which should be the standard in a high quality journal like ACP.
- **Inferred Na⁺:** The other major issue is the use of "Inferred Na⁺", which I am still uncomfortable with. First, "Inferred Na⁺" should be renamed since it accounts for all NVCs, not just sodium. Perhaps "Inferred NVCs"? Secondly, as one of the reviewers noted, inferred Na⁺ is a bit of a circular argument and somewhat misleading. In prior work, the authors' have argued that proxy methods, such as molar ratio and ion balance have significant shortcomings, but to my understanding "inferred Na⁺" uses the same basic concept since inferred Na⁺ is used to help explain the ammonium-to-sulfate ratios, even when the actual Na⁺ data don't match that. Maybe this could be reframed with a different name to help the issue. However, introducing a fudge factor and implying it has a real meaning (e.g. Na⁺ concentration) to account for the fact that the measurements of Na⁺ (which have their own issues) don't match the inferred value necessary to have NVCs explain the R ratio, is not strong support for the authors' hypothesis and argument that NVCs explain the ammonium-sulfate ratio.
- **Exclusion of Organic Films, Glassy Aerosols, or other Possibilities:** The abstract and paper overall still very much imply that the authors' hypothesis about NVCs is the only reasonable explanation for overprediction of R values. However, given the issues with the Na⁺ data (below LOD) and the need to use inferred Na⁺ values higher than what is measured to make the hypothesis work, the strength of the authors' arguments against organic properties/films or other explanations does not appear sufficiently justified and should be weakened (as noted in previously).