Comment on amt-2021-327
Anonymous Referee #2

This paper describes a series of experiments looking at the amount of organic impurities in ammonium sulfate solutions and also across different particle sizes when those solutions have been aerosolised. The authors present a convincing argument for a significant size dependence on the ratio of [Org]/[sulfate] and have suggested the results indicate that the organic is concentrated at the surface of the particles rather than forming an internally mixed particle. This is a highly relevant and interesting result, which has important implications for laboratory simulations and measurements of physiochemical properties. I recommend publication after some minor corrections.

Line 82: Organosulfates can be formed via a range of routes, not only by isoprene epoxides. I would suggest adding a few more examples and references.

Line 88: It would be helpful for the reader to know a bit more about how ammonium sulfate is manufactured. This would allow some indication of the potential sources of organic contaminants.

Line 90: is the 0.8 % in the ammonium sulfate without humic acid? Its is a bit confusing what is meant here.

Line 115: I would like some more details about the air/N2 difference. Zeroi air is also likely to contain organics and it is important to determine whether the air is also a source
Need to explain that the second two are relative to the NO3.

What is the difference in the chromatograms using the two different eluents – you say its been optimised but I don’t see any discussion about why one set was used over another. Also, why is only +ve mode used for optimisation? This may result in you discounting negative mode as unimportant, when it hasn’t actually been optimised specifically.

“slightly higher (respectively lower)” – this is unclear and I don’t follow the meaning.

what is the source of the nitrate ions?

“large organic molecules”? The AMS doesn’t provide justification for this statement. The LC-MS may, but I don’t think this statement is justified here. Also, what does "large" mean?

While not outside your uncertainties, there certainly seems to be a pattern in the effect of concentration on the ratio. Can this data be included in the SI?

what does “multi-characterization” mean here?

I would change the colour scheme here – I got confused with the standard AMS colour scheme i.e. red = sulfate.

Give the DBE for the neutral masses not the ions.

This sentence doesn’t make sense. Needs reworded.

Is this the TIC or base peak chromatogram? I assume the later due to the quality of the chromatogram.
Line 334 and earlier – To me direct injection means injection of a solution directly into the MS source without prior chromatography. This needs to clarified.

Line 340: I think “mark” should be “make”

Line 339: no data is presented to back up the “factor or 20 lower” – can you include a chromatogram in the SI?

Line 355: Is the mass accuracy really good enough to assign a C24 peak?

Line 379 – add the particle size to this final statement.

Line 394-396: This needs to be reworded – it seems like one sentence split into 3