Review of "Retrieval of the Sea Spray Aerosol Mode from Submicron Particle Size Distributions and Supermicron Scattering during LASIC", by J. Dedrick, et al., submitted to Atmospheric Chemistry and Physics.

This study used the particle number size distribution from UHSAS and scattering coefficients at 3 wavelengths from Nephelometer to construct the size distribution of sea spray aerosols. This topic is of great scientific interest. However, I have some concerns about the method, evaluation, and presentation of this study, before it can be considered for publication.

Major:

- 1. The instruments in LASIC, i.e. Neph, UHSAS, PSAP, are not performed under dry conditions, while at different RHs. I have three main concerns regarding the comparability of the different parameters either measured or calculated in this study:
  - The mean RH of Neph is ~60% while UHSAS is ~55%, so this method is actually constraining the Mie simulated scattering with the measurements under different RHs. How will the RH difference influence the results? The scattering coefficient can be quite different for aerosols at 55% and 60% RH due to hygroscopic growth, especially for the strongly hydrophilic sea spray aerosols. The 5% RH can not be simply ignored just because "the uncertainty of the RH values (~10%) likely falls within the mean and range of reported humidity for each instrument" (page 5 line 150 to 154). Since this is the basis of this approach, more discussions and evaluations concerning the RH difference between Neph and UHSAS are needed to make this study robust and convincing.
  - 2) This study used 1.56+0i from previous studies for the refractive index, while the refractive index also changes with RH, and eventually influence the simulated scattering coefficients. Authors should take it into consideration in the Mie calculation.
  - 3) Authors compared SSA with those in previous studies; please make sure these values are comparable. The SSA in this study is not under dry conditions and might be overestimated as a result of the enhanced scattering coefficients due to aerosol hygroscopic growth.
  - Section 4 authors stated "supermicron scattering was independent of the instrument RH up to a values of approximately 60%", while it showed an apparent increase of scattering coefficients with RH from 50% to 66% RH in Fig. S5.
- 2. In the evaluation section, we do not notice any significant improvements of the UHSAS-Neph method compared to the UHSAS-only method, except for the correlation between scattering coefficients with the sea spray particle mass (Fig. 9c and 9d). However, this correlation should be good for the UHSAS-Neph method as authors are using the scattering to constrain sea spray particle size distribution. Besides the good correlation with scattering, the UHSAS-Neph method is only slightly better (0.2 vs 0.1 for R) than the performance of the UHSAS-only method in the correlation with wind speed and worse in the correlation with the chloride mass

concentration of submicrometer particles. I would recommend moving S2 to the main context to help evaluate this method.

- 3. In-situ measurements found dust is one of the most observed aerosols at Ascension Island (Schenkels, 2018; Swap et al., 1996). I cannot tell how dust is differentiated from sea spray aerosols in this study. Please clarify.
- 4. The last paragraph of the introduction gives a detailed description of the method, which should be moved to the method part. The second paragraph of the introduction should be expanded to give a general review of previous studies. In particular, this study compared the results with those from the UHSAS-only method, which should definitely be introduced in the introduction.
- 5. I think this paper is missing a finalizing step. There are lots of grammar, citation, and reference format mistakes (e.g. line 49, line 434, line 470, line 472, and line 686). As there are lots of native speakers in the author list, this should not be a problem. I strongly suggest authors to do a thorough reading of this paper.
- 6. The authors used the range of "shoulder" to predict sea-spray number size distribution. However, this range is not consistent throughout the paper. In Fig. 1, it shows 0.1-0.4  $\mu$ m; while it changed to 0.4-1  $\mu$ m in line 124, and to 0.38-0.9  $\mu$ m in line 389, and again to 0.38-0.83  $\mu$ m in line 392. The authors need to explain the variation of the shoulder range.
- 7. Nucleation mode, Aitken mode, accumulation mode, and coarse mode are four modes used to describe the aerosol number size distribution. This paper used a lot of "sea spray mode" or "sea spray aerosol mode", which is not common usage, I suggest authors to revise.
- 8. In Section 3.4, the authors converted number size distribution to mass size distribution by assuming the density to be 1g cm<sup>-3</sup> for sea spray aerosols. This value seems to be quite low, less than half of the value of 2.017g cm<sup>-3</sup> used in Zieger et al. (2017). Please clarify the reasons for choosing this density.

## Others:

- 1. Page 3 line 64, please clarify "relative availability".
- 2. Line 144, again, the Neph is not measured under dry conditions, I strongly suggest not to use "dry scattering coefficients".
- 3. Line 166, the citation "Dmt, 2017" is incorrect. Please revise according to the journal's citation and reference format.
- 4. I would suggest adding the ranges of  $\sigma_{sca,1-10\mu m}$  and  $\sigma_{PNSD,mea}$  in Table 2.
- 5. Please clarify "MAOS" in line 181.
- 6. I suggest authors to specify under which RHs different variables are measured in Table 1.
- 7. The threshold of 0.07 ppb for CO seems extremely low (line 229 and Fig. 3).
- 8. Please add references in line 271, 314, 519, and 522.
- 9. I would suggest moving S1 to the main context.

- 10. Authors use a lot of terms in MATLAB, e.g. normrn, I would suggest avoiding using them or adding explanations for these terms.
- 11. Please clarify the "low error-restricted Mie solution".
- 12. Please explain why "These high fit RSS likely indicate that the supermicron scattering measurements were influenced by particles other than sea spray" in line 397 and "had fit RSS values above 5, possibly because they may have been influenced by dust or other non-marine aerosol intrusions that were not effectively screened using the clean marine criteria" in line 404.
- 13. Line 400, please add a table in the supplement showing the variation of correlations with different thresholds.
- 14. Please clarify "no observations during this period" in line 419.
- 15. Please clarify the location of the description for the UHSAS-only method in line 469.
- 16. The value in line 483 is inconsistent with the one in Table 4.
- 17. In the evaluation section, the authors stated that the sigma from the UHSAS-only method is much broader than those from the UHSAS-Neph method; please clarify which sample from UHSAS-Neph is using here; the 1237 or the 971? If it is the 971 one, would the constraints in Section 4 help reduce the sigma range and result in the narrower range?
- 18. The discussion of Fig. 8 needs revision. In line 533, the authors stated  $\sigma_g < 2$ , while Fig. 8b is actually >2. No discussion for Fig. 8e?
- 19. In Fig. 9, the authors use  $R^2$  for Fig. 9c and 9d, while R for others; why?

Schenkels, M.: Aerosol Optical Depth and Cloud Parameters from Ascension Island retrieved with a UV-depolarisation Lidar; An outlook on the validation, Master thesis, Utrecht University, 2018.

Swap, R., Garstang, M., Macko, S. A. et al: The long-range transport of southern African aerosols to the tropical South Atlantic, J. Geophys. Res. Atmospheres, 101, 23777–23791, https://doi.org/10.1029/95JD01049, 1996.

Zieger, P., Väisänen, O., Corbin, J. et al: Revising the hygroscopicity of inorganic sea salt particles. Nat Commun 8, 15883, <u>https://doi.org/10.1038/ncomms15883</u>, 2017.