

Interactive comment on “Dry versus wet marine particle optical properties: RH dependence of depolarization ratio, backscatter and extinction from multiwavelength lidar measurements during SALTRACE” by Moritz Haarig et al.

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I have read with great interest your discussion paper on the hygroscopic effects of aerosol optical properties of marine aerosol particles. A few remarks came up during the reading of the manuscript which might be helpful.

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Classification of 'dry particles' and the effect of relative humidity on the scattering enhancement factor

The question of when a particle can be considered 'dry' is a tricky one. Thresholds of relative humidity (RH) < 50 % are mentioned within the manuscript. This value is not sufficiently low to determine that a particle is dry for a number of reasons. Firstly, as has been shown in numerous studies, sea spray aerosol particles take up water at relatively low RH's (see e.g. Fig. 1 in Tang et al. (1997) or Fig. 2 in Zieger et al. (2017)). Even at RH's close to 0 % the inorganic sea salt particles will still contain water, due to the presence of hydrates which will influence their overall hygroscopic growth (Zieger et al., 2017).

In addition, the history of the RH the particles have experienced is important. If particles have experienced high values of RH, that is an RH above their main deliquescence RH, which is most likely the case if they were freshly formed in the marine boundary layer, they will stay on the upper branch of the hysteresis curve down to roughly RH ≈ 40 %.

To better demonstrate this effect, I have plotted in Fig. 1 the calculated scattering enhancement factor $f(\text{RH})$ of pure inorganic sea salt based on measurements conducted at Stockholm University using a sea spray simulation chamber (Salter et al., 2014). The calculations were performed using Mie theory based on measured size distributions (Salter et al., 2015) and the recently determined hygroscopic growth factors (Zieger et al., 2017) as input. At RH ≈ 50 %, the remaining water can contribute up to a factor of 2 to the scattering compared to dry conditions. But even if the particles have experienced very low RH, the contribution of water can be up to 20-30 % of the overall particle light scattering coefficient. Therefore, careful consideration of the temporal evolution of the RH that particles have experienced is critical. Again, this highlights the point that a threshold of RH = 50 % is too high to classify particles as 'dry'.

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The effect of particle shape

Inorganic sea salt is a complex mixture of inorganic salts and includes hydrates. This complex composition, along with the rate at which the particles have dried (see Wang et al., 2010), has implications for the shape of the 'dry' sea salt particles. As we have recently shown, inorganic sea salt particles tend to be more spherical than pure NaCl especially with increasing particle diameter (see Fig. 1 in Zieger et al., 2017). Indeed, even pure NaCl particles are not always perfect cubes (Zelenyuk et al., 2006; Zieger et al., 2017). These points should also be borne in mind.

A few more specific comments are listed below:

- Page 1, Line 19: Maybe this is a wrong reference, Tang et al. (1997) did not look specifically at the shape of sea salt particles.
- For the DDA modelling (Sect. 3.3), it would be helpful to know the assumed size parameters (mode diameters, width, etc.).
- Page 8, Line 28: I would not use the term 'water shell' for a marine aerosol particles since the majority of the chemical components (i.e. inorganic salts) will be dissolved at elevated RH.
- Page 8, Line 30: You will never be 100 % sure that only marine aerosol was present. I would add the word 'mainly' before 'marine aerosol'.
- Page 9, Line 2: Similar to the comment above, you can never fully exclude other aerosol sources although they are very unlikely. I would soften the language here.
- First paragraph on page 10: The discussion on the RH-dependency of the LR could be expanded by relating the presented observations to previous literature (e.g. Ackermann, 1998; Zieger et al., 2011).

- Page 10, line 27: Here, I would make it more clear that you have observed a specific case of the dehydration of particles along the upper branch of the hysteresis curve (i.e. that they have fully deliquesced before they are dehydrated again). In addition, one should keep in mind that inorganic sea salt has multiple efflorescence points due to its complex composition (see e.g. Tang et al., 1997; Zieger et al., 2017).
- Equation 2 and Fig. 13: The upper and lower branches of the hysteresis curve would have to be fitted separately. Therefore, one would not expect a value of $A = 1$ but rather a value of $A > 1$ for sea salt aerosol, especially if you consider that most of the particles will have fully deliquesced at one point and be on the upper branch of the hysteresis curve. Since the values of A are here clearly below 1, I assume that the reference RH of the backscattering coefficient is not sufficiently low (or just not known).
- Equation 2 and Fig. 13: Here, you should also discuss that these results are only valid if the reference RH of 40 – 50 % is sufficient to represent ‘dry’ particles, which, as discussed above, is probably not the case. This will be a critical point for modellers who will relate their ‘wet’ values of aerosol optical properties to perfectly dry values (at RH=0%). As such, they will subsequently estimate much higher enhancement factors than shown in Table 2 or Fig. 13. Figure 1 below shows the same γ -fit as used in the presented study. Extrapolating to RH=0% will give a value of approx. $A \approx 1.5$ (with $\gamma \approx 0.45$).

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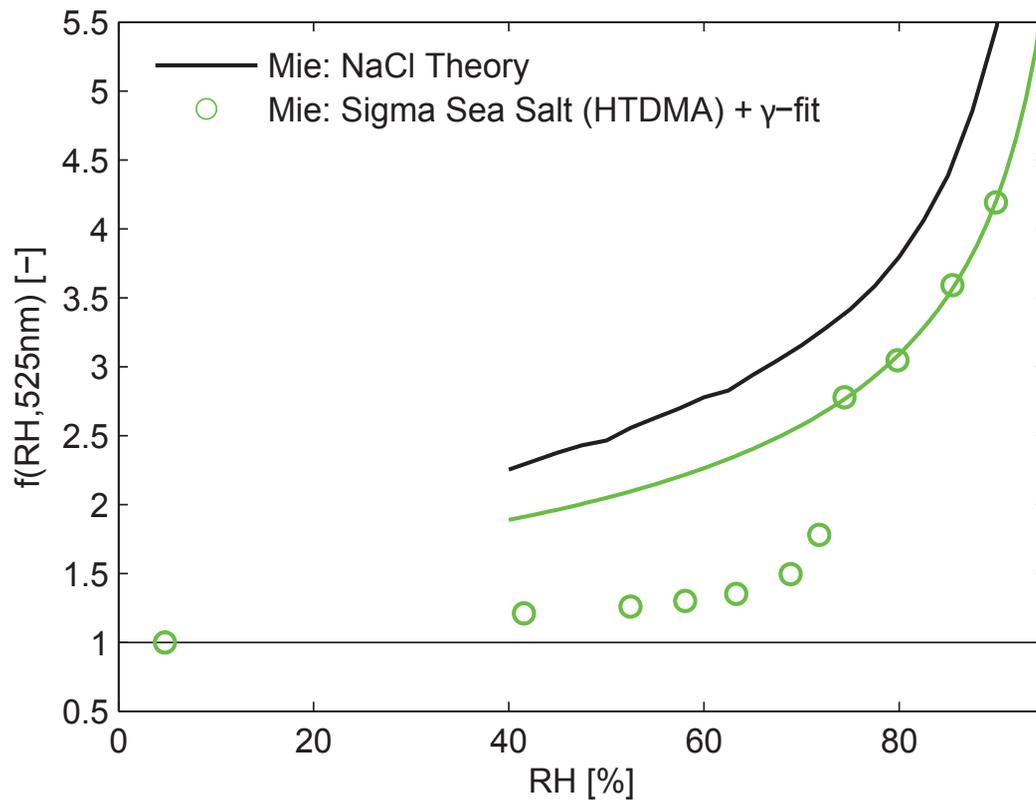


Fig. 1. Scattering enhancement of inorganic sea salt calculated using Mie theory (see text for details).

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