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Interactive comment on "Establishing the Impact of Model Surfactants on Cloud Condensation Nuclei Activity of Sea Spray Aerosols" *by* Sara D. Forestieri et al.

Anonymous Referee #3

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This manuscript presents a high quality experimental and modeling study of the effect of coatings of organic surfactants on monodisperse NaCl particles. This is a relevant and timely piece of work adding interesting insight to the discussion of the role of surfactants in cloud droplet activation and explaining literature observations that highly surface active fatty acids have little effect on CCN activity.

I enjoyed reading this manuscript and recommend publication after minor revision.

Comments and suggestions

Abstract: in the abstract it is stated that a kinetic limitation to water uptake is observed for NaCl particles coated with pure palmitic acid, I suggest the coating thickness is

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Discussion paper

provided.

Introduction

Page 4: There are several studies relevant on NaCl, surfactants and ccn activity showing different effects which could be included in the discussion – e.g. Petters and Petters J. Geophys. Res. Atmos.121 , 1878, King et al. Environ. Sci. Technol. 2012, 46, 10405. The different results obtained illustrate the importance of a better understanding of surfactants.

Methods Page 5: line 23. It should be specified (given in nm) what is meant by "thin" and "thick" coatings respectively.

Description of coating experiments: I suggest to include a schematic of the flow tube and associated dilution system in Figure 1.

How can the residence time of 0 s be understood? That would be injection and sampling at the same spot? Why is that discussed?

Perhaps I misunderstand something, but if the concentration of ozone is 278 ppb in the flow tube and the pressure is atmospheric pressure (∼2.46x10ˆ19 molecules per cc) how can the ozone exposure be so high as 10ˆ14 molecules per cc? (278*10-9*2.46*10ˆ19 ∼ 7x10ˆ12 molecules per cc) – is there a dilution effect involved?

Regarding calibration: reviewer 1 has already raised the subject that kappa values are not constant with particle size and I agree that this should be addressed in the revised manuscript. In their reply the authors have shown that this is not relevant for their particles sizes for NaCl. What about ammonium sulfate? On a related note shape factors of 1.08 and 1.04 are included in the calibration of the CFSTGC – is it based on the literature or have shape factors for the specific system used by the authors been measured?

Was a shape factor also applied in the calibration of the CCN counter? This should be stated.

It would be nice to see a figure with the particle size distributions before and after coating – in some cases the coatings are very thick - how broad were the size distributions after coating?

Some references for the equations given and the underlying assumptions would be nice, e.g. equation (6) is it assumed that all organics are at the surface? Relation between equation 6 and 10 –somewhat overlapping but have different symbols (A and A_mic). I don't think the symbol for molecular volume of the organic is explained in relation to equation 10.

It is a bit difficult to get an overview of the experiments actually performed. In Table 1 it just says the type of coating. How many different dry sizes, RH values and coating thicknesses form the basis for each set of fitted parameters?

As I understand f_surf was an assumed parameter - I miss a discussion on the importance of f_surf and what values of f_surf were assumed in relation to the parameters Table 1.

Perhaps I have overlooked it, but I could not find quantitative information on the magnitude of the uncertainly on RH – it says "Uncertainty in the film model curve was estimated by perturbing the input RH values by the average precision-based uncertainty in RH." – what was the actual average precision-based uncertainly in RH?

Linking to sea spray aerosol and secondary marine aerosol: I find the discussion a bit hard to follow – what is meant when referring to "high" CCN activation efficiency, it says kappa_app >0.7 – but 0.7 is lower than the kappa for artificial sea salt which is around 0.9? Some literature references should be given to support the statement "The often large CCN activation efficiency for particles observed in the marine environment, in particular nascent SSA particles". Perhaps also references to work showing other trends should be included; for comparison, it has been reported that particles generated from sea water or sea water spiked with sea surface microlayer had CCN activity similar to sea salt (Rasmussen et al. 2017, J. Aerosol Sci. 107, 134) and high kappa values in ambient environment can also be related to organic-to-sulfate ratio (Yakobi-Hancock et al. 2014 , Atmos. Chem. Phys., 14, 12307). Since also aged and secondary particles are addressed, it could also be mentioned that surfactants may come from the gas phase (Sareen et al. Proc Natl Acad Sci U S A. 2013, 110, 272).

Minor Title: The title says “Sea spray aerosols” – I suggest changing to “model sea spray”, “salts” or “Sea spray aerosol mimics” or similar since the study was performed on NaCl core particles.

In the caption for Table 1 caption it says: “Uncertainties on the film model parameters on the precision in RH (see text for details)” – it is not clear to me what is meant here.

Figure S2: it should say in the figure caption that the calibration was performed with NaCl particles.

Figure 2: figure text- “confidence interval” – is this 95% ? should be stated.

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