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## *Interactive comment on* "Properties of cloud condensation nuclei (CCN) in the trade wind marine boundary layer of the Eastern Caribbean Sea" *by* T. B. Kristensen et al.

## Anonymous Referee #1

Received and published: 2 December 2015

The manuscript "Properties of cloud condensation nuclei (CCN) in the trade wind marine boundary layer of the Eastern Caribbean Sea" by Kristensen et al. presents the results of a month-long campaign in Barbados focusing on the CCN properties of the ambient aerosol. The paper discusses on-line particle measurements, including CCN number concentrations and aerosol particle size distributions, as well as the CCN parameters derived from these measurements. The presented work is further supplemented by an off-line particle analysis utilising transmission electron microscopy and energy-dispersive X-ray spectroscopy techniques. In general, the paper is a wellrounded overview of the CCN properties in the marine environment in question; it is well-written and contains new and exciting information in the field of ambient CCN

C9983

studies. I recommend this paper for the publication in the Atmospheric Chemistry and Physics journal after the following general comments and technical corrections have been considered.

## General comments

1. At the moment the main concern of the manuscript is exactly why there is not as much sea salt aerosol (and hence, low kappa values) as one would expect from a marine environment location. The authors say that the wind direction was from the ocean sector for > 95% of the time, and the top panel in Fig. 1 shows that the wind speed was predominantly above 6 m s–1 throughout the campaign. All begging the question: where was the sea salt? CCN in marine environments have been investigated in detail in published literature, and authors do compare their results to other campaigns in the region. However, at the moment, the discussion as to why exactly sea salt concentrations were so low and such low kappa values were observed is insufficient. An unsuspecting reader would be very surprised to see kappa values of 0.3 across the whole size range for a location dominated by marine air masses.

I recommend to expand sections 5.2 and/or 6 to include the discussion about the abovementioned apparent discrepancy. What was so different about the meteorological conditions (besides the wind speed) during the first day of measurements? Do the references of (Monahan and Muircheartaigh, 1980; O'Dowd and de Leeuw, 2007) provide any numerical estimates of the fluxes of sea salt particles as a function of wind speed, and how do the presented data compare to those numbers? Was anything different about the instrumental setup during the first day of measurements? Has anybody previously reported such low hygroscopicity in the marine environment? Was the Köhler equation used correctly (see minor comment #7 below)? Since the presented results are that different from the published literature (both experimental and modelling), it is important to provide enough detail as to why that is the case.

2. While the authors do compare their results to several other published studies, it

might improve the paper to put the results in the global perspective using any of the many overview papers discussing CCN properties at a multitude of locations around the world (including marine). CCN concentrations can be compared to e.g. Andreae (2009), while CCN concentrations and hygroscopicity can be compared to e.g. Paramonov et al. (2015).

Minor and technical comments

1. While the paper is well-written, its quality and clarity can be greatly improved with the proper use of punctuation. This refers to the use of connector words, the word "respectively" and sentences consisting of two or more clauses.

2. Similarly, please, be consistent with the use of British English or American English (e.g. sulphate vs. sulfate).

3. Make sure that all instances of the noun "data" in the text reflect its plurality.

4. Please, consider the use of past tense when talking about the experiments you conducted. Section 4.2 is one instance where the present tense is used in several sentences.

5. page 30759, line 1 – change the (IPCC, 2013) reference to (Boucher et al., 2013)

6. When discussing the marine organic compounds and their effect on CCN hygroscopicity (as, for example, on page 30760, 2nd paragraph and on page 30776, 2nd paragraph), a useful reference could be Ovadnevaite et al. (2011).

7. page 30761, equation 1 – what exactly is SS in the equation? If SS is supersaturation in %, it should be possible to enter e.g. 0.7%; this, however, is incorrect as it will lead to an error. Please, refer to Equation A(30) in Rose et al. (2008) for the correct equation.

8. page 30763, line 12 – the units of the flow rate should be volume per time. Please, correct

C9985

9. page 30764, lines 7-10 - "a critical diameter (Dc) above which all particles activate into cloud droplets for a given supersaturation". This is true only if the aerosol is internally mixed. Please, modify

10. page 30766, lines 16-19 - the sentence is not very clear; please modify

11. page 30767, lines 12-17 – the wind direction that is used to delineate the northern/northwestern boundary between the open ocean and the local sources is used inconsistently throughout the text ( $335^{\circ}$  vs.  $360^{\circ}$ ). In text on page 30767 it says that Fig. 1 uses the  $360^{\circ}$  boundary, the caption of Fig. 1, however, mentions the  $335^{\circ}$ boundary. Please, fix. Also, lines 15-17 on this page can be removed from the text as they should be in the figure caption (see comment 1 for the Figures).

12. page 30768, lines 3-6 – this sentence belongs in the figure caption, not in the text.

13. page 30768, lines 24-25 – the end of the sentence after the comma should read ", and, thus, has not been carried out".

14. page 30770, lines 20-22 – chemical composition and potentially particle sources change with the seasons? It's probably likely that both chemical composition and particle sources vary throughout the year. Please, consider this and check the reference.

15. page 30771, lines 22-24 – this statement is only true for the time when the experiments were conducted (June and July). It is not possible to say whether this would be true at all other times of the year, since coarse particles may have different chemical composition throughout the year (see page 30770, lines 20-22 and the comment above). Please, modify

16. page 30773, lines 18-19 – "where the last category contains all refractory substances not belonging to any of the first mentioned groups". Such as?

17. On two different instances (page 30774, lines 15-18 and page 30776, lines 19-20) the paper mentions that the investigated aerosol can be considered to be representative of the (Eastern) Caribbean Sea. This is technically not true for two reasons. First,

the geographical eastern boundary of the Caribbean Sea goes along the line of Lesser Antilles, of which Barbados is already the easternmost island. Second, it is mentioned in the text that during > 95% of the measurement time the wind direction was from the  $0^{\circ}$  to  $130^{\circ}$  sector (the ocean), meaning that the aerosol did not, in principle, spend any time at all above the Caribbean Sea. The investigated aerosol is, therefore, more likely representative of the Atlantic Ocean (or the westernmost tropical Atlantic, to be completely precise), with influence from long-range transport, as mentioned in the text. Please, modify.

18. page 30774, line 26 – "was" should be changed to "were"

19. page 30774, lines 26-27 continuing to the next page 30775, lines 1-7 – the authors first state that during most of the campaign the mass concentrations of mineral dust were significant. The authors then go further and say that the bulk of the particles was dominated by non-refractory species and that the concentration of refractory PM was minor. This is confusing and somewhat contradictory. The authors should either clarify what they mean by "significant" when talking about mass concentrations of mineral dust or replace "significant" with a more appropriate adjective.

20. page 30776, lines 2-4 – while the chemical composition can be comparable, the kappa values are most certainly not. Reconsider

21. page 30776, line 29 – please, fix the reference parentheses

22. page 30777, line 1 – should read "...emissions are the main source..."

Figures and Tables

1. Fig. 1 – for the mid panel (wind direction), please, extend the y-axis to 360°. The figure caption does not say what the horizontal dashed line is. If the boundary of 335° is correct (see minor comment #11 above), also include the horizontal line for this wind direction. Shading instead of the grey vertical dashed lines may also improve the figure.

2. Fig. 2 – the first sentence of the figure caption should read "Size distributions C9987

corrected neither for particle losses in the tubing nor for detection efficiency". OR "Size distributions not corrected for particle losses in the tubing or for detection efficiency". Also, to improve the detail, the upper panel of Fig. 2 can be wider, if possible. The label of the colour bar must include (cm–3).

3. Fig. 3 - is there a reason that the data at SS of 0.3% are missing in the upper panel?

4. Fig. 6 – in the caption for panels (a) and (b), there is no need to describe the refractory classes and the associated colours. This information is easily visible in the legend. Defining rf here is sufficient.

5. Table 1 – in the caption, are presented kappa values averages or medians? Also, please, mention in the text (section 3) that the CCNC measured at the SS of 0.3% only during the last 15 days of the campaign.

## References

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organic aerosol: A dichotomy of low hygroscopicity and high CCN activity, Geophys. Res. Lett., 38, L21806, doi:10.1029/2011GL048869, 2011.

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- Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, Atmos. Chem. Phys., 8, 1153–1179, 2008.

Thank you for an excellent paper!

C9989

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 30757, 2015.