

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.

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We would very much like to thank Referee 1 for the excellent and detailed comments related to the manuscript. We find that the changes suggested by Referee 1 have improved the paper significantly.

Below we address the Referee 1 comments in a point-by-point fashion.

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1 General Comments

1. Referee 1 finds that in comparison to the literature the reported κ_{CCN} values are unexpectedly low, and the influence from sea salt is surprisingly low. The CCN activity results presented in the manuscript provide information related to the mobility particle diameter size range from ~ 50 to ~ 200 nm. Based on hygroscopicity measurements for that size range available in the literature from the marine boundary layer (MBL), it appears that the hygroscopicity typically is dominated by inorganic sulphate species with a potential significant influence from organic species and occasionally a moderate influence from sea salt (Swietlicki et al., 2008) with the most pronounced hygroscopicity mode being represented by a κ value of ~ 0.45 (Wex et al., 2010). Pringle et al. (2010) modelled the CCN activity in the MBL in the region where our measurements were carried out. They predicted a κ value of 0.6–0.7 in winter and a κ value of ~ 0.4 in summer, which fits very well with our reported results from the summer period and the previous measurements in the region carried out during winter (Allan et al., 2008; Hudson, 2007; Pringle et al., 2010). Hence, our reported CCN activities are very much in line with what could be expected from various previous measurements and model studies. Furthermore, our results agree with the reported chemical composition of particulate matter in the relevant size range in the region being dominated by inorganic sulphate and organic species (Novakov et al., 1997).

The text has been modified accordingly to make this agreement with previous findings more clear. A comparison to the modelled κ is included (see below under 2) (Pringle et al., 2010). The following has been added after P30770L22: "Wex et al. (2010) reviewed measurements of particle hygroscopicity for sub-saturated conditions in the MBL and found that the dominant κ value was around 0.45, but also several observations with lower κ values and fewer with higher κ values have been made. Hence, our inferred κ values are slightly lower than what most frequently can be expected in the remote MBL."

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Answers to specific questions raised by the Referee:

During the first day of measurements - nothing but the maximum in wind speed stands out when it comes to the local meteorology. Hence, it may be that the indication of an enhanced fraction of sea salt was to a larger extent produced up-wind on this particular day.

O'Dowd and de Leeuw (2007) mention that sea salt can reach a maximum number concentration in the MBL of 50-150 cm^{-3} under high wind conditions. Of course much higher wind speeds than the observed ~ 14 m/s can be encountered in the MBL, so it is likely that the number concentration of sea salt in general is much lower in our case - as also indicated by our findings.

The instrumental set-up was identical for the entire time period covered in the manuscript.

The Köhler equation was used correctly - as shown below.

To sum up: Our findings of a negligible influence from sea salt in the diameter range from ~ 50 to ~ 200 nm agrees very well with most previous findings in the MBL. It is likely that relatively higher wind speeds are needed for a significant fraction of sea salt particles in that size range.

2. The CCN number concentration has been compared to results reviewed by Andreae (2009) , and the inferred κ values have been compared to the model results presented by Pringle et al.(2010).

The following sentence has been added after P30769L14: "Andreae (2009) reviewed measurements of CCN number concentrations at an SS=0.4% in the 'clean' MBL and reported an average concentration of $107 \pm 56 \text{ cm}^{-3}$, with slightly higher values for the

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Atlantic. Hence, the median CCN number concentration of 205 cm^{-3} we observed for an SS=0.4% is in the higher range for the remote marine MBL."

The following sentence has been added after P30770L22: "Pringle et al.(2010) modelled the κ values in the region to have a maximum of ~ 0.7 in winter with a κ value of ~ 0.4 in summer, which agrees very well with our findings."

2 Minor and Technical Comments

1. Commas have been added in the following lines: P30759L05, P30770L21

2. "Sulfate" has been changed to "sulphate" in P30759L09 and P30766L23

3. The plurality of data is now reflected in the following lines:

In P30771L19: 'data was' -> 'data were'. In P30772L08: 'data provides' -> 'data provide'. In P30787 Fig. 2 caption line 2+3: 'data covers' -> 'data cover'.

4. Past tense has been introduced in the following lines:

P30766L14: "is" -> "was". P30766L24: "remain" -> "remained". P30766L25: "can conclude" -> "concluded". P30767L01: "is" -> "was"

5. The reference has been changed from IPCC (2013) to Boucher et al.(2013) as suggested.

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6. Ovadnevaite et al.(2011) do not report the CCN activity of the studied aerosol particles, and their qualitative arguments related to the CCN activity does not account for differences in particle number size distributions. The focus of their study is primary marine organic compounds. The organic compounds present in the current study are likely to be of secondary origin (Mayol-Bracero et al., 2001; Novakov et al., 1997). Thus, we do not find that there is a firm basis for comparing our findings to what is reported in the suggested study.

7. SS in the equation is the supersaturation in %. It is indeed possible to use Eq. (1) with e.g. an SS=0.7%, so It is not quite clear which error in Eq. (1) the Referee refers to. If eq. A2 and eq. A30 from Rose et al.(2008) are combined, then Eq. (1) is obtained as presented in the manuscript.

From Rose et al.(2008):

$$S = (s - 1) \cdot 100\% \quad (A2)$$

$$s = \frac{D_{wet}^3 - D_s^3}{D_{wet}^3 - D_s^3(1 - \kappa)} \exp\left(\frac{4\sigma M_w}{RT\rho_w D_{wet}}\right) \quad (A30)$$

where S is the supersaturation in percent and s is the saturation ratio in their notation. The remaining parameters are as defined in the Kristensen et al. manuscript - with a few differences in notation. By combining the equations (A2) and (A30) from Rose et al.(2008) the following equation is obtained - which is identical to Eq. (1) presented in the manuscript:

$$\frac{SS}{[100\%]} = \frac{D_d^3 - D_p^3}{D_d^3 - D_p^3(1 - \kappa)} \exp\left(\frac{4\sigma M_w}{RT\rho_w D_d}\right) - 1 \quad (1)$$

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8. In P30763L12: The flow rate unit has been corrected to $\sim 40 \text{ m}^3/\text{hr}$.

9. The sentence in P30764L07-10 has been modified to: "For an estimation of the CCN activity, the average CCN number concentration was compared to the simultaneously measured particle number size distribution. Thus, a critical diameter (D_c) above which all particles activate into cloud droplets for a given supersaturation was estimated assuming internally mixed aerosol particles in a sufficient neighborhood around the inferred D_c ."

10. The sentence in P30766L16-19 has been rephrased from: "As during particle analysis, the volatile material of the particle evaporates and the refractory fraction remains, images were recorded at the beginning of the electron bombardment and after no more visible change in particle structure occurred." to: "During the electron beam analysis, the particles absorbed energy and heated up. Hence, the volatile particulate material evaporated and only the refractory fraction remained on the substrate. To determine the original state, one image was recorded right at the beginning of the electron bombardment. When no more change of the remaining particle fraction was observed, a second image was recorded. Changes to the particles were derived from image comparison."

11. The 360 has been changed to 335 in P30767L16. P30767L15-17 have been removed.

12. The P30768L03-06 have been deleted - similar information is available in the figure caption.

13. The text has been modified as suggested.

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14. The sentence in P30770L20-22 has been modified to: "It is worth noting that the previous measurements were carried out in December and January respectively, and it is likely that the PM sources and chemical composition change with the seasons".

15. Yes, we agree. The following expression was added to P30771L22-24: 'during this campaign, where mineral dust mass concentrations were significant.'

16. An example of a refractory particle not classified as 'soot', 'soot-mixture', 'dust', nor 'sea-salt' is shown in Fig. 5 V.b. As can be seen from Fig. 6.c+d: unidentified refractory particles did not comprise a significant volume fraction. The following sentence has been added after P30773L19: 'An example of a refractory particle classified as "other" is shown in Fig. 5V.b'.

17. The expression in P30774L17 has been changed from: "representative for the Eastern Caribbean Sea" to: "representative for the trade wind MBL of the Western North Atlantic."

As a consequence of this referee-comment/modification several other corrections have been carried out:

The title has been changed from: "Properties of cloud condensation nuclei (CCN) in the trade wind marine boundary layer of the Eastern Caribbean Sea" to "Properties of cloud condensation nuclei (CCN) in the trade wind marine boundary layer of the Western North Atlantic"

The running title has been changed from: "CCN Caribbean" to: "CCN in the trade wind MBL of the Atlantic"

In the abstract, P30758L05: "Eastern Caribbean" -> "Western North Atlantic".

P30759L27-28: "Eastern Caribbean" -> "Western North Atlantic". P30777L14: "in the

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trade wind marine boundary layer in the Eastern Caribbean Sea" -> "in the trade wind marine boundary layer in the Western North Atlantic"

In P30776L the statement about the location does not only refer to our presented data but also to the LIDAR measurements carried out by Groß et al.(2015) on the west coast of Barbados. So "Mineral dust was observed over the Caribbean Sea" has been changed to: "Mineral dust was observed over the Eastern Caribbean Sea/Western North Atlantic".

18. P30774L26: "was" -> "were" as suggested.

19. The fact that very small mineral dust concentrations are observed in the accumulation mode by use of TEM is not necessarily contradictory to significant dust mass concentrations being present simultaneously - since the mineral dust appears to dominate the supermicron size range during the campaign. To make that more clear - the following sentence: "The majority of the investigated particles were found in the accumulation mode, and in general the bulk of the particles were largely dominated by non-refractory substances." has been extended to: "The majority of the investigated particles were found in the accumulation mode, and in general the bulk of the investigated particles were largely dominated by non-refractory substances. Those findings indicate that the mineral dust primarily was present in the larger particle size ranges in the MBL during the campaign."

20. Novakov et al.(1997) and companion studies related to the properties of organic species in aerosol particles on/near Puerto Rico did not report any direct measurement of the CCN activity since the applied method/approach is indirect and associated with significant potential errors (Novakov and Penner, 1993; Novakov et al., 1994). So it is not quite clear if the κ value of the organic species in the previous studies indeed

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would be substantially higher than e.g. 0.08. Since our comment only is related to the organic fraction - we do not find that it is misleading.

21. The reference parenthesis has been modified as suggested.

22. The text has been modified as suggested.

3 Figures and Tables

1. The y-axis of the mid-panel has been extended to 360 as suggested, and the boundary of 335 has also been indicated by a horizontal dashed red line. The vertical dashed lines have been replaced by shading with a more light gray colour. The figure caption has been modified accordingly as suggested.

2. The first sentence of the caption has been modified as suggested.

3. The reason for excluding the data at SS=0.3% was graphical and not due to any scientific reasoning. The data at SS=0.3% are now included in the figure for completeness.

4. The figure caption for Fig. 6 has been shortened as suggested.

5. The presented κ values are medians. The caption has been modified as suggested. In P30763L03 the following has been added: "Supersaturation settings of 0.1%,

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0.2%, 0.4%, and 0.7% were applied throughout the campaign, and an SS=0.3% was included during the last 2 weeks of the campaign."

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