Atmospheric
Chemistry
and Physics

Discussions

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 #2

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The paper presents cloud condensation nuclei concentrations together with aerosol particle number size distributions and off-line particle analysis as observed in the marine boundary layer (MBL) in the Eastern Caribbean (Barbados) in June/July 2013. With this set of instrumentation the authors wanted to assess how organic species from marine biological activity, long-range transported mineral dust, and sea salt particles affect the particles' ability to act as CCN since in the past several contradicting observations have been made for MBL aerosol. Better knowledge of aerosol-cloud interaction, however, is necessary for the correct incorporation of clouds in climate models, especially those in the MBL. Interestingly no significant number or volume fractions of sea salt or dust could be observed in the sub-micron range, which is the

relevant size for CCN activation. Thus the inferred hygroscopicity parameter, kappa, can be only explained by a mixture of sulfate and organics.

The paper is well structured and discusses the results nicely. The information given in this manuscript is relevant for the readers of ACP and of interest for the community in general. I recommend the paper for publication in ACP after the following comments and corrections have been considered by the authors.

General comments:

1. The authors mention that all the in-situ measurements were carried out through a PM10 inlet located at the top of a 17 m high tower. In contrast, the dust mass was obtained from a high-volume sampler collecting total PM and the TEM samples were taken with a cascade impactor with nominal cut-sizes of 530 nm to 90 nm or 330 nm to 90 nm, respectively. There are two things, which should be considered. Firstly, all instruments cover different size ranges. This should be emphasized a bit more in the paper by adding the size ranges at several places in the text (see following specific comments). Secondly, different line losses are probably involved in the measurements. Samples for off-line analysis were collected directly at the top of the tower, whereas the inlet for the online instruments was located on the top of the tower with the instruments placed on the ground level. It is written that no correction for coarse particles in the sampling lines was made because it is "uncertain". I do not understand what the authors mean with "uncertain". If they know all the tube lengths, bends and flow rates they should be able to calculate the losses (von der Weiden et al., 2009). I think that this correction is very important since one expects to have relatively high numbers of coarse particles for the air masses involving mineral dust and sea salt and since there was a quite long way from the inlet to the instruments. Indeed, the gravitational settling in a (hopefully mostly) vertical line are small, but already for short horizontal lines losses of particles larger than 1μm are quite substantial. If the correction for line losses were included, it would affect the estimated PM but maybe also the derived kappa. In contrast, the offline samples should be, I guess, not affected by substantial losses. I

do not know in which way the sampling lines were installed during the campaign, so maybe the effect is only little, but I think the authors should discuss in more detail in their paper what the effect of losses can be and how that translates in the comparability of the individual measurements.

2. On page 30763, the authors write that "to minimize impacts from local sources sampling was controlled by a wind sensor and only carried out when the local wind direction was in the range 335° . . . to 130°, and the wind speed exceeded 1 m s-1". This refers to the high-volume sampling, but does this also apply to the impactor sampling and to the size distribution and kappa averaging? It does not say so in the paper. If not, what would be the expected difference of data points within this wind pattern compared to the rest of the data points? Would not the influence of local sources be more significant for Aitken and accumulation particles (the main size range of TEM samples, CCN and size distribution measurements). Would it make sense to exclude times with local influence for the size distributions, CCN properties and TEM analysis?

3. The authors motivate their study with the need for better understanding of aerosol-cloud interaction in order to incorporate MBL cloud development into climate models. My question is whether it can be expected that the observed CCN properties also prevail at higher altitudes in the atmosphere, i.e., at levels where clouds form? What if there is a dense long-range transported dust layer at higher altitude including large numbers of coarse particles? They would all be activated into cloud droplets no matter what their hygroscopicity is. Please compare the study by Jung et al. (2013), who report significantly increased CCN concentrations in a Saharan dust layer at ∼2.2 km altitude compared to ground levels. It would be nice if the authors could discuss the effect of strongly changing aerosol load with height on cloud formation.

Specific comments/ Technical corrections:

TEM samples: On p. 30763 is written that there were two time periods of sampling

for which different size ranges were analyzed. The different size range is an important point and should be mentioned not only in Sect. 3 but also in Tab. 2, Fig. 6, and/or Sect. 5.4. How much can the projected area equivalent diameter (PAED) differ from the aerodynamic diameter? I thought that in general they should be more or less equal. I am wondering because for period 2 the TEM samples have quite high number fractions for PAED > 0.3 μm (higher than for period 1) although the samples were only analyzed for impactor stages up to 330 nm.

Discussion on measured kappa (starting on p. 30775, l. 24): This paragraph tells a lot of estimation on how the measured kappa values could be explained. What I am missing is a bit more detailed explanation on the calculations. For example, it is written that assuming internally mixed species of organics and sulfate kappa is explained assuming 50% organic volume fraction with kappa = 0.08 and 50% sulfate with kappa = 0.6. I understand that this can be calculated from Eq. 2. But in another sentence is written that if an external mixture of sulfate and organics is assumed, an organic volume fraction of 25% would explain the observed kappa. Please make clear how this can be calculated.

p. 30762, l. 22-25: Please add the respective size ranges that are covered by the individual instruments.

p. 30764, l. 7-10: Which number size distribution was integrated – the one obtained by MPSS or by MPSS and APS? From the text following it sounds like only the MPSS size distribution was integrated. Please clarify. Since the CCN counter was attached to the PM10 inlet, it would also activate particles larger than 0.8 μm. There might be not many particles > 0.8 μm but still I guess it would make more sense to calculate the critical diameter from integrating the full (MPSS + APS) size distribution. This should lead also to a smaller kappa.

p. 30763, l. 12: "flow rate of 1m3" per which time?

p. 30765, 28: I do not understand what the "(SS = 0.074 %)" tells.

p. 30773, l. 9: add "of" before "the super-micron"

p. 30774, l. 26: change "was" to "were"

p. 30775, l. 9: ammonium sulphate in two words

p. 30776, l. 1: "if it is..." change "it" to "is"

p. 30776, l. 29: put the reference in brackets

p. 30777, l. 10: add "of" before "organic species"

Fig. 2: It would be more convenient to display the color bar as real concentration values and not as the logarithm of the concentration. Also, it would be better to distinguish between individual days, if the grid lines were on top of the colored surface plot.

Fig. 3: Please add the CCN concentration at SS = 0.3% to Fig. 3a. Would it make sense to plot the concentration and Dc on a logarithmic scale?

Fig. 4: Please add the particle size range that is included in the dust mass and PM_est, respectively.

Fig. 5 a and b: What happened to the blue sea salt bar? It appears several times in one PAED group and is always behind the other colored bars. Should it not appear as a separate bar next to the others?

References:

Jung, E., B. Albrecht, J. M. Prospero, H. H. Jonsson, and S. M. Kreidenweis (2013), Vertical structure of aerosols, temperature, and moisture associated with an intense African dust event observed over the eastern Caribbean, J. Geophys. Res. Atmos., 118, 4623–4643, doi:10.1002/jgrd.50352.

von der Weiden, S.-L., Drewnick, F., and Borrmann, S.: Particle Loss Calculator – a new software tool for the assessment of the performance of aerosol inlet systems, Atmos. Meas. Tech., 2, 479-494, doi:10.5194/amt-2-479-2009, 2009.

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