

Dear Reviewer,

We thank you for doing this review and for your suggestions that helped to improve our manuscript. Below, please find your original comments in blue and our responses in black. When referencing page and line numbers, we are always referring to the original versions of manuscript and SI.

The manuscript discusses aerosol size distributions and CCN measurements made at multiple supersaturations observed at Cape Verde as part of the MarParCloud project. Measurements were conducted at two stations – a low-altitude, coastal station (CVAO) and a higher-altitude, mountainous station (MV), with the latter station being located close to cloud level. The size distributions are fit to 2-3 lognormal modes (Aitken, accumulation, coarse), and very rough inferences of aerosol type are drawn from relative fractional contributions of these modes as well as HYSPLIT air mass backtrajectories. CCN measurements show size-dependent hygroscopicity with lower kappas observed at higher supersaturations (with the implication that these measurements are representative of smaller aerosol sizes). This leads to the interesting conclusion that cloud processing both transitions aerosol from the Aitken to accumulation mode, but also slightly increases the kappa. Overall, the paper is well written and relevant to ACP. I recommend publication after the following comments are satisfactorily addressed:

1) The paper as it is currently written stands on its own, and from the brief description given of the second paper, it also sounds like it too will adequately stand on its own. Consequently, I recommend that the title be revised so that this is not be a two-part paper. Alternatively, the authors should provide a copy of the companion manuscript and explain why the two are inextricably linked.

The two manuscripts are inextricably connected to each other. Part 2 is under discussion on ACPD. Please refer to this link for more details. <https://www.atmos-chem-phys-discuss.net/acp-2019-729/>

The two companion manuscripts both belong to the MarParCloud project and discuss the aerosol present at Cape Verde from 13 September to 13 October, where both manuscripts examine data from the CVAO at sea level and from the Monte Verde. Part 1 focuses on aerosol particle number size distribution (PNSD) and CCN number concentration and Part 2 focuses on ice nucleating particles (INPs). Part 2 does somewhat rely on Part 1, as it is important that we observed, in Part

1, that the marine boundary layer is well mixed, which cannot be deduced for INP (due to the cloud events, which only enable to get a few INP filter samples from Monte Verde). We would therefore prefer to keep the titles as they are now.

2) The language and concept of deploying a "trimodal parameterization method" as described in the abstract and elsewhere (e.g., Pg. 9, Lines 2-3; Pg. 24, Lines 7-8) implies that something novel has been developed, which is not the case. The size distribution measurements are fit to multiple lognormal functions to derive summary statistical parameters, using fit functions that are textbook and commonplace. Please revise this language to indicate that the "parameterization method" is actually "fitting the data to multiple lognormal functions".

The "trimodal parameterization method" is a well-known method to fit the particle number size distribution. We cited the textbook (Seinfeld and Pandis, 2016) in the manuscript.

For clarification, we changed page 9, lines 2-3 to:

"A well-known trimodal log-normal parameterization method is adopted to characterize the temporal variation of PNC in three modes."

We changed page 10, lines 10-11 to:

"To better define the modes of our data, we fitted the PNSDs to three log-normal functions."

3) How were the size modes and backtrajectory information synthesized to arrive at the four aerosol type classifications in the present paper? Would it make more sense to conform to the 5-type classification scheme of Fomba et al. (2014)?

The particle classification was based on the particle number concentration in different modes. The classification criteria was summarized in Tab. 2. The backward trajectories (1-hour resolution) were used for characterizing the different particle sources. In other word, the backward trajectories were used to double check if our particle classification was reasonable or not. Theoretically, if we classified the particle sources based on backward trajectories and checked the particle size distribution for different sources, the results should be similar to this paper. We do think the PNSD

measurements are more precise than the backward trajectories; therefore, we used the PNSD to do particle classification rather than used the backward trajectories.

Fomba et al. (2014) classified the particle sources from 2007 to 2011 at CVAO according to the backward trajectories (24-hour resolution) and then discussed the chemical composition difference for different particle sources. This manuscript focused on particle number concentration and Fomba et al. (2014) focused on particle mass concentration. Fomba et al. (2014) carried out 5-year measurement, which means more aerosol types were found. For example, Fomba et al. (2014) found that the PM<sub>10</sub> could reach ~200 µg/m<sup>-3</sup> during heavy dust periods, while in this study, the PM<sub>10</sub> was at most about ~40 µg/m<sup>-3</sup> during dust type 2 period. A comparison of particle classification between mass concentration and number concentration is discussed in the MarParCloud project overview paper (submitted to ACPD).

4) The sentence on Pg. 2, Line 14 is awkward and unnecessary. I suggest it be removed.

Done.

5) Pg. 2, Ln. 22-23: Karydis et al. (2011) did not find that dust contributes up to 40% to CCN on a global basis. This was found for the N. African and Asian desert regions.

Thanks for your comment. It was changed to:

“Karydis et al. (2011) found that the predicted annual average contribution of insoluble mineral dust to CCN number concentration in cloud forming areas is up to 40% over North Africa and Asia (Arabian Peninsula and Gobi Desert).”

6) Pg. 3, Ln. 9: remove "besides"

Done.

7) Pg. 3, Ln. 12: Quinn et al. (2017) did not find that "marine aerosol" contributes less than 30% to CCN. They use the term "sea spray aerosol", and suggest that SSA contributed less than 30% to CCN. Organics and secondary sulfate of marine origin can dominate CCN in remote regions.

The first reviewer suggested defining "marine aerosol" in the manuscript. We add the following in page 3, line 4:

"Together with newly formed particles originating from gaseous precursors which can also be emitted from the ocean, this sea spray aerosol (SSA) contributes to marine aerosols.

Therefore, in page 3, line 12, we changed it to:

"On a global basis, SSA makes a contribution of less than 30% to the CCN population (Quinn et al., 2017)."

8) Pg. 3, Ln. 16: Something is amiss with the total mass reported of  $47.2 \pm 55.5$ , as it implies substantial negative mass ( $\sim -8.3 \text{ ug/m}^3$ ). I suspect that the observations here lack normality and the use of an arithmetic mean and standard deviation is inappropriate.

Yes, you are right. The standard deviation is larger than the mean, implying that the data is not normally distributed for a strictly positive data set. It would be better to use median and interquartile range to represent of the data. However, only mean, median and standard deviation were reported in Fomba et al. (2014).

9) Pg. 3, Ln. 31 (and multiple instances elsewhere): The use of the phrase "to the best of our knowledge,..." is sloppy writing and gives the reader the impressions that the authors have not done their due diligence in conducting a literature survey. If the statement is true (which I think it is), then it should stand on its own without the need for such a caveat.

We removed "To the best of our knowledge" in the manuscript.

10) Pg. 3, Ln. 31: "filed" = "field"

Done.

11) Pg. 4, Ln. 18: "see" = "sea"

Done.

12) Pg. 4, Ln. 20-21: Is it really the first time these measurements have been conducted in Cape Verde? Why is the "to the best of our knowledge" caveat here?

We removed "To the best of our knowledge" in the manuscript.

13) Pg. 4, Ln. 25: Please update reference or remove it if the paper is still in preparation.

The overview paper is submitted to ACPD. This information will be updated in the new version.

14) Pg. 4, Ln. 30-31: Are the winds always from the northeast?

Yes, they are. The Cape Verde islands receive north-easterly prevailing trade winds blowing directly off the ocean. This is described in Carpenter et al. (2010).

15) Pg. 4, Ln. 31-33: Please add citations to support these sentences related to annual rainfall and precipitation even frequency.

Done. We cited Fomba et al. (2013) and Carpenter et al. (2010).

16) Pg. 7, Ln. 3-4: How was the APS data used to correct the MPSS data for multiple charges as the APS is measurement aerodynamic diameter? What assumptions were invoked?

The dry density of Saharan dust particles was determined in a range of  $\rho = 2450 - 2700 \text{ kg m}^{-3}$  over the Cape Verde Islands (Haywood et al., 2001). The dry particle density of sodium chloride is known to be  $\rho = 2160 \text{ kg m}^{-3}$ . The overall effective density of the dust and sea-salt fraction is approximately 2, as recommended in Schladitz et al. (2011).

The dry dynamic shape factor  $\chi$  of mineral dust is  $\chi = 1.25$  (Kaaden et al., 2009) for 1  $\mu\text{m}$  particles, whereas the dynamic shape factor for sodium chloride is  $\chi = 1.08$  (Kelly and McMurry, 1992; Gysel et al., 2002). We used the average shape factor of 1.17 in this study.

Based on these, a conversion from aerodynamic to geometric diameters were done for the APS data, and particle number concentrations from the APS were used to correct the multiply charged particle concentrations in the upper size range where the MPSS measured.

Above information will be mentioned in the supplement.

17) Pg. 7, Ln. 5: "base" = "basis"

Done.

18) Pg. 7, Ln 11: Please add a sentence to the end of this paragraph summarizing how approximately how large the particle loss corrections ended up being (e.g., on the order of 10%, something smaller, or something larger?).

We added: "Overall, less than 3% of the particles were lost when passing the inlet."

19) Pg. 10, Ln. 2: What is meant by "behavior of aerosols" here? Is this discussed in this manuscript?

This might have been a misleading formulation and we removed it such that the sentence now is:

"Particle size is one of the most important parameters to characterize the atmospheric aerosol."

Table 1: Please reformat the table so the Measurement Site and Location fields are on the same line as the other information.

Table 1 was changed.

Table 3: I don't understand what is being presented in the kappa column. Is one of the numbers the + and the other the -? If so, which is which. Would it be better to report the geomean \*/ geostd?

It is the geometric standard deviation (geostd). The first number means the geomean+geostd and the second means geomean-geostd. We changed the explanation in the table as geomean, +geostd, -geostd and reported the three numbers. It should be clear now.

Figure 12: It would be really interesting to use the median size distributions from Fig.5 to compute and overlay lines of constant kappa for each case to evaluate how the box-whiskers fall across the range of hygroscopicities.

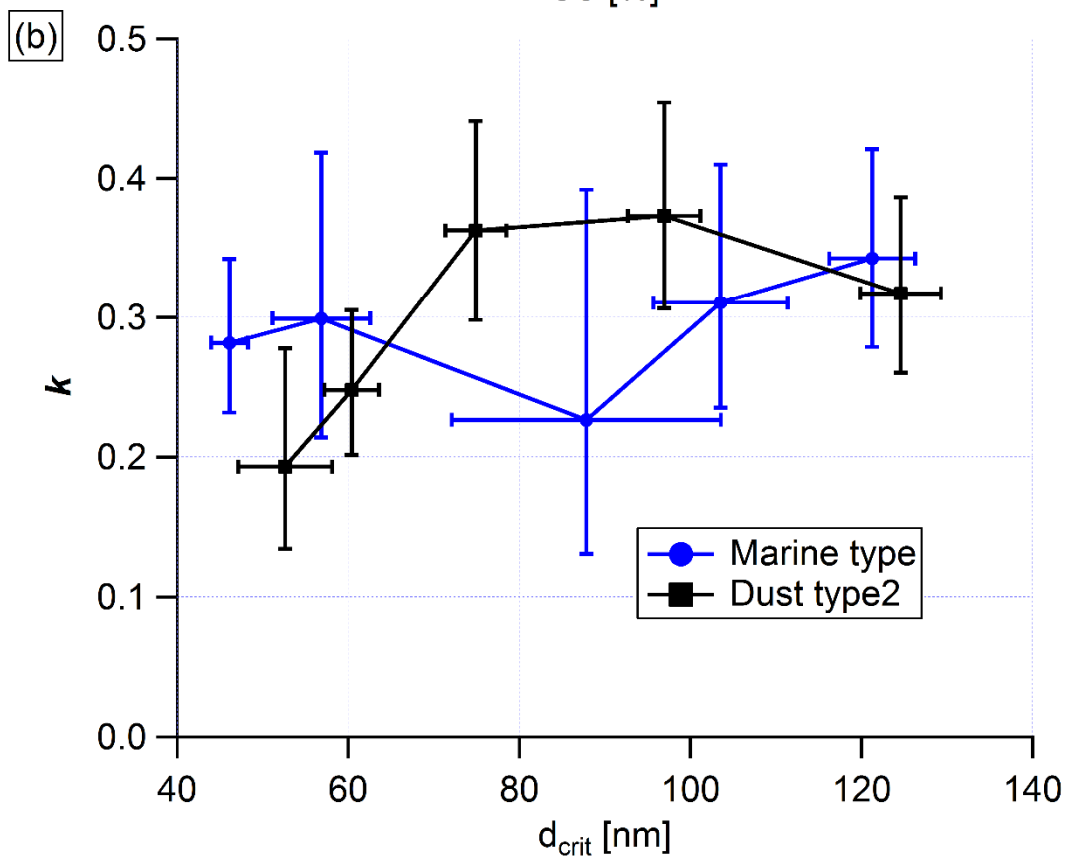
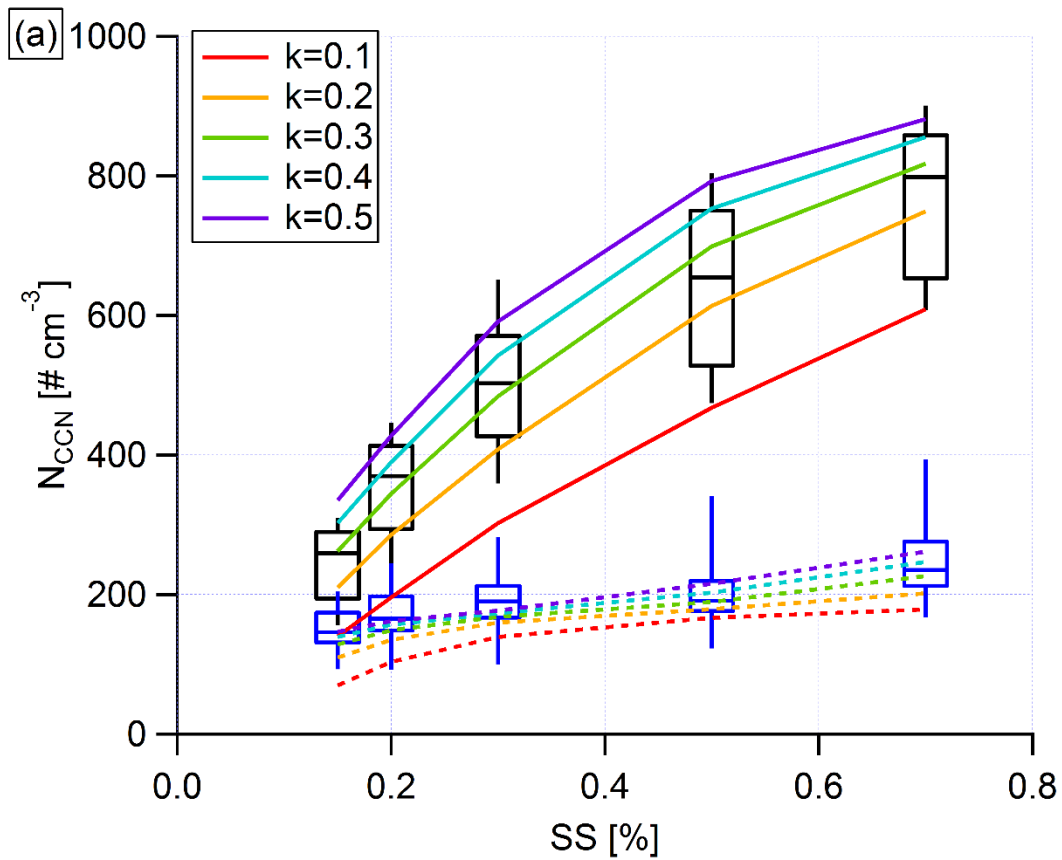
We are sorry that it is not totally clear to us what you mean. We assumed you want to see the calculated CCN number concentration by assuming a certain  $\kappa$  value, based on the measured median PNSD.

We added the following in page 23, line 2:

“We additionally derived  $N_{CCN}$  based on PNSDs. For that, we assumed values for  $\kappa$  of 0.1, 0.2, 0.3, 0.4 or 0.5, and calculated the corresponding  $d_{crit}$  at different supersaturations. The integrated particle number concentration in the size range larger than  $d_{crit}$  were derived from the median PNSDs during dust type2 and marine periods. These particle number concentrations also can be treated as the predicted  $N_{CCN}$  at different supersaturations, as shown in solid (dust type2) and dashed (marine type) lines with different color (indicating different  $\kappa$ ) in Fig. 12(a). As expected, the thus derived  $N_{CCN}$  were within the measured  $N_{CCN}$  range. Comparing the solid and dashed lines, it can be seen that different aerosol types, i.e., different PNSDs, played an important role in  $N_{CCN}$  variation. When looking at the results from the different  $\kappa$  values, we found the particle chemical composition did not control  $N_{CCN}$ , especially when the particle number concentration was very low. These colorful solid and dashed lines connected the  $\kappa$  and  $N_{CCN}$ , which can be helpful for  $N_{CCN}$  predictions in modeling studies.”

The title of Fig. 12 was changed to:

“(a) Boxplot of  $N_{CCN}$  as a function of  $\kappa$  for marine type (blue) and dust type2 (black). Whiskers show the 10% to 90% percentiles. The predicted  $N_{CCN}$  based on median PNSD and different  $\kappa$  values (0.1, 0.2, 0.3, 0.4 and 0.5) are shown in solid (during dust type2 period) and dashed lines (during marine period). (b)  $\kappa$  as a function of  $d_{crit}$  for marine type (blue) and dust type2 (black). Error bars of  $d_{crit}$  and  $\kappa$  show 1 standard deviation and 1 geometric standard deviation, respectively.”





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