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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "CCN concentration and INP-relevant aerosol profiles in the Saharan Air Layer over Barbados from polarization lidar and airborne in situ measurements" *by* M. Haarig et al.

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General Comments

This paper continues a series of papers themed around the use of lidar retrievals to estimate CCN and INP profiles. These developments of capabilities are being followed with interest by a broad community since the applications and utility are obvious for regions where in situ data are not available, or not available with high frequency. This paper will make a nice incremental contribution to the growing literature base of this team, focusing here on comparison to aircraft data that did not have INP data to compare to. I do have some critical comments and suggestions in a few regards.

1) First, I believe that the nature of retrieval of multiple species contributions to aerosol number, mass, CCN and INP requires some additional description because this is a more recent development (versus retrievals from layers dominated by a single aerosol type) and so bears reiterating from its introduction over the last two years. If the Marinou et al. paper is accepted for publication, reference should be made to the detailing of the detailed schematic there.

2) Secondly, I feel that the use of the DeMott et al. (2010) parameterization as specific to continental and non-dust contributions to INPs is not exactly correct, and this has implications. The "continental" definition in Mamouri and Ansmann (2016) neglects the fact that dust contributions to INPs were most certainly folded into the parameterization in a variety of environments. I see now that Marinou et al. (2018) have written, "As the majority of the samples used for D10 are non-desert continental aerosols, this INP parameterization has been considered to be suitable for addressing the immersion and condensation freezing activity of mixtures of anthropogenic haze, biomass burning smoke, biological particles, soil and road dust (Mamouri and Ansmann, 2016)." This is also a gross simplification, with the actual contributors unknown, and the likelihood that dust was folded in at a variety of levels of contribution. After all, one study was PACDEX, the Pacific Dust Experiment. Hence, strong caveats about potential duplication of INPs, and lack of assured attribution to all of the other types mentioned, are needed here. What one may really wish for are parameterizations for all relevant INP species instead. Substitution of D10 for the absence of such detailed information is not ideal, and so I am concerned that this is being glossed over. It is worrisome that this assumption seems to have propagated into a number of papers since 2016, and in some cases is even called "non-dust" or continental "pollution", the former not being true to the original paper and the latter being a true stretch in attribution that has never been supported by direct evidence.

3) I also wondered about the use of the groups' own parameterization of sea spray aerosols (based on DeMott et al., 2016, since that paper did not promote a direct

parameterization) versus a marine-specific parameterization for the Atlantic region that is referenced in the introduction (McCluskey et al., 2018). Do they compare well? I obviously know the answer, but you might justify persisting with a parameterization that did not as deeply consider "pure" marine as did the newer McCluskey paper. I realize that this is a very minor point, since marine INPs at -25 °C are minor contributors compared to mineral dusts in SAL conditions.

4) For the use of the DeMott et al. (2015) parameterization (D15), it seems that a decision has been made to not use the recommended 3x correction factor for immersion freezing that was justified in that paper? If so, the basis/reasoning for this should be stated.

5) Finally, I think that it would be very useful to demonstrate retrievals in a profile that does not necessarily include dust or smoke overlying or mixing in the region above the marine boundary layer. That would represent the unperturbed case, and give insights into the behavior of the combined set of parameterizations when dust is not at all dominant.

Additional context to these comments and some additional specific questions/editorial comments for addressing before publication are listed below.

Specific Comments

1) Page 2, lines 2-4: What papers are you referring to in stating the implementation of these parameterization schemes? These are not all included in this present paper, although it would be interesting to see. Also, please note that there is no parameterization given in DeMott et al. (2016). This must have been created by the authors.

2) Page 2, line 10: fix "several 10000 km" to state a range of distances expected.

3) Page 2, lines 25-27: Note that as written, the sentence is repetitive in mentioning dust and smoke mixture at the beginning and ends of the sentence.

4) Page 3, lines 2: The continental aerosol designation is not mentioned here, as listed

in Table 2. As stated above, this needs some serious caveats applied, namely that it is used in the absence of a true set of parameterizations that could describe other than mineral dust input, even though it definitely includes some influence from varied levels of mineral dusts in the studies used by D10. It was not intended to be specific or neglectful of any particular class of INPs.

5) Last paragraph of Page 3, and start of Page 4: This discussion of assumptions on the hygroscopicity of mineral dust wanders some and never quite makes clear if kappa values for Saharan dust after transport to the region have been measured as low as is assumed or if this is an assumption based on the "fresh" nature of dust observed via say, microscopy studies. There is a difference, as trace amounts of materials can make a difference. In the end, it seems that the value selected of 0.02 is in the range of most measurements (i.e., not fully hydrophobic), and in the range estimated to be consistent with activation in clouds as submicron dust particles in the Eastern Atlantic (Twohy et al., GRL, 36, L01807, doi:10.1029/2008GL035846, 2009).

6) Page 4, line 14: The statement "The very hydrophilic sea salt particles (sodium chloride) have an activation diameter…" sounds awkward. Sea salt is hygroscopic. But sea salt is rarely the composition of sea spray particles alone, so why not say that "We assume a composition of sea salt for marine aerosols, and prescribe an activation diameter of…"

7) Page 5, line 23: Perhaps discuss that cumuli attenuate the lidar, versus "disturbed" the measurements?

8) Page 5, paragraph starting line 29: This is where I suggest that some elaboration on the methods for retrieving the contributions of different aerosols in a mixed scenario is given.

9) Page 6, line 21: the CCN data from the Falcon are "measurements." They may have uncertainties, but they are not retrievals.

10) Page 7, first paragraph: Is the surface area used only for the marine parameterization? Do the dust parameterizations using s significantly differ from D15? I only wondered about the derivation of surface area if it was not going to be used.

11) Page 7, lines 8-10: This is a rather subjective statement about the likely role of the INP concentrations derived for the SAL. Clearly, direct cloud observations or cloud model simulations are likely needed to explore the implications, since such tropical cumuli are known to contain rather vigorous secondary ice formation processes through their deep supercooled layers (e.g., see Lasher-Trapp et al., J. Atmos. Sci., 73, 2547-2564, 2016, and references to Lawson et al., 2015 and Heymsfield and Willis, 2014 therein).

12) Page 7, line 21: Consider replacing "Wrong in situ particle counting. . ." with uncertainties in in situ aerosol measurements. There is no support provided for how or why the measurements would be wrong. They are your only link to apparent ground truth.

13) Page 8, line 15: suggest leading to "likely changes in trade wind cumulus cloud microphysical properties. . ." rather than "developments". Also, does it not depend on which layer dominates aerosol contributions to convective clouds?

14) Page 8, line 32: Suggest "reconciled" for "fixed"

15) Page 9: The summary paragraph is a bit short in its outlook for the future. You would seem to benefit from more validation INP data, particularly for cases with and without dust, so the validity of the apparent knowledge of continental that you promote is also checked. And not only in dusty situations. Will you have INP data in any of the forthcoming campaigns? Quantifying other specific aerosol type contributions than dust and marine would appear useful as well.

16) Table 1 header: the cv coefficients need explanation. Are these the "conversion factors" mentioned?

17) Table 2: as mentioned, DeMott et al. (2016) does not include a parameterization, so

that is not the appropriate reference for it. I suggest using the specific parameterization of McCluskey et al., if that is possible. Also the D15d reference needs to mention somewhere (if not in the table) what cf factor is used in this study.

18) Figure 4: I can note similar s values here as in DeMott et al. (2015) for the SAL over the Western Caribbean, but the predicted INP concentrations are a bit lower here at around -25°C. This motivated me to ask about the cf factor assumed for use in the parameterization in the present study. Also, below 2km, is it certain you are dealing with dust and not marine aerosols in all cases? Is this why the lidar profile showing higher surface area and n250 on 22 June still leads to a decrease in D15-predicted INPs? Is that because you presume all of those particles are "continental"? This is where I think the application of a D10+D15 approach could lead to errors, and the only way to tell will be future in situ INP measurements.

19) Figure 6 inspired me to ask what an unperturbed profile might look like, for example when there is not a strong dust or smoke or pollution layer over the clouds. Do you have any such data?

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