

We thank Reviewer 1 for their constructive comments. We reproduce reviewer comments in *blue* in the following.

Page (P.) 2, line (L.) 16: The reference Klein et al. (2010) may be omitted, because of serious experimental errors in these data, as stated in a recent paper by this laboratory (Schrod et al., Atmos. Meas. Tech., 9, 1313–1324, 2016).

We agree and have deleted this reference.

P.2, L. 20: Huang et al., JGR 115, 2010 give a nice climatology of dust AOD over the subtropical atlantic from MODIS, you might add it as a reference here.

We have added the reference.

P.3, L.20: Conen et al. (2015) measured immersion freezing nuclei, this could be stated.

We have added “immersion mode” before “INP concentrations at 265 K” on P.3, L.22 of the revised manuscript.

P.4, L. 19: Uptake of water, deliquescence and growth begin below water saturation. Assuming 100% as a threshold is reasonable for practical reasons, as we usually have nothing better for a given situation, but it is not the truth.

We have replaced P. 4, L.19-21

“Above water saturation condensation freezing, where ice starts forming while water vapor condenses on an INP, as well as immersion freezing, where the INP is immersed in a droplet prior to initiating freezing, were investigated. The latter two processes cannot be distinguished with our method and are thus only referred to as condensation freezing.”

with P.4, L. 19-22 of the revised manuscript:

“Close to and above water saturation condensation freezing, where ice starts forming while water vapor condenses on an INP, as well as immersion freezing, where the INP is immersed in a droplet prior to initiating freezing, were investigated. The different processes cannot be distinguished with our method and thus we refer to deposition nucleation at $RH_w < 100\%$ and to condensation freezing at $RH_w \geq 100\%$.”

P.7, L.5: I recommend to replace “dried” by “evaporated” or “sublimated”. I presume that all ice is completely evaporated once a particle enters WIBS?

Yes, this is the case. We have replaced “dried” by “evaporated” (now P.7, L.13)

P.7, Chapter 2.5: I wonder if a flow scheme would help a reader who is not familiar with your setup, but I realize that it can be looked up in your JAS paper.”

We have added a flow scheme for the standard set-up and the coupling of PINC-PCVI and WIBS. This is the new Figure 1 on P.26

P.5 ,L.2-3 : *“A schematic of the experimental set-up is given in Fig. 1.”* was added.

P.7, L. 10: *“An overview of the coupled set-up is given in the right panel of Fig. 1.”* was added.

P.8,L.6: Do you have more details or a reference on how gains and losses were accounted for ?

We have added the following sentence on P.8, L. 11-13:

“As described in the Appendix of Boose et al. (2016) a size-dependent loss curve of the impactor was measured using montmorillonite and Arizona Test Dust. The size-dependent enrichment of the concentrator was determined using Arizona Test Dust. These loss and gain terms were multiplied with the aerosol particle size distributions.”

P.10, 3.3, Fig.4 and various other places: when R2 or R are compared, the number N of observations is often useful.

We have added the number of observations, “ n_{obs} ” to all R and R² values, including those in Table 3.

P. 10, L. 5-10: To my knowledge the first one to publish a correlation of INP to the number concentration of “large” particles (0.1-1 μm dp) were Georgii and Kleinjung (Journ. de Recherches Atmosphérique, 145-156, 1967). This reference may be added.

Georgii and Kleinjung (1967) was added on p. 10, L. 15.

P.13, L.1-2: since you excluded the (blue) squares from the Atlantic sector in your analysis of Fig. 9c you should write “... collected in the SAL under Saharan influence...”. In the same sense you could add, that in Fig. 9d there is no correlation for the red symbols (for red and blue together one might get the impression that there is a correlation).

We have added *“under Saharan influence”* on P. 13, L.18

We have further added *“... for the Saharan samples (red circles in Fig. 10d).”* on p.13, L. 19-20

P.13, last sentence: It is not plausible to use Fig. 9f as an example for the large scattering and variability, because this is the plot that is least scattered of all the 6 plots in Fig.9, less than others that you use for interpretation.

We do not intend to use Fig. 9f as an example for large scattering. We agree that the scattering is the lowest here and this makes sense. Actually, if fragmentation took place in a constant manner, one would expect no scattering in this plot at all since with more dust mass the number of dust particles should increase linearly. Therefore, the sentence describes that the fact that there is some scattering is due to the dust agglomerate fragmentation.

To clarify this, we have appended (P.13, L- 32-35 original manuscript):

“As shown in Fig. 5h, the surface area alone could not fully explain the differences in observed [INP]. This indicates that the degree of fragmentation of the dust agglomerates (Perlwitz et al., 2015) influences the

variability of the number of INP. The scattering of the $N_{0.5-1\ \mu\text{m}}$ to dust_1 plot (Fig. 10f) illustrates the variability in the dust agglomerates fragmentation.”

as follows (P.14, L.14-18 in the revised manuscript):

“As shown in Fig. 6h, the surface area alone could not fully explain the differences in observed [INP]. This indicates that the degree of fragmentation of the dust agglomerates (Perlwitz et al., 2015) influences the variability of the number of INP. If fragmentation was constant, a linear relationship between $N_{0.5-1\ \mu\text{m}}$ and the dust_1 mass would be expected. The scattering of the $N_{0.5-1\ \mu\text{m}}$ to dust_1 plot (Fig. 10f) illustrates thus the variability in the dust agglomerates fragmentation.”

P. 14: I suggest “Potential sampling bias” as a header of chapter 3.7

The header of chapter 3.7 on P.14 has been changed to “Potential sampling bias”.

P. 15, L. 22-23: The parameters that are displayed in Fig.’s 12b and d could be described more clearly . “observed “ is misleading, because it is more “what would have been observed, if there were no gains and losses”. Also in the sentence “Figure 12b and d show ambient concentrations“ the term “ambient” might be understood in this way, one could add, that it is derived from measurements.

We have changed the respective sentences on p. 15, L. 22-24 (original manuscript)

“Figure 12b and d show ambient concentrations. Here, the observed [INP] include the omitted INPs, as described in the previous section. Thus, for the predicted [INP] the ambient size distribution of particles between $0.5 \leq d_{ve} \leq 20\ \mu\text{m}$ was used without further corrections.”

to p. 16, L. 8-13 (in the current manuscript):

“Figure 13b and d refer to ambient concentrations. The [INP] displayed on the x-axes are those measured with PINC and corrected for the omitted INPs, as described in the previous section. Error bars include the Poisson error of the measured [INP], 10 % uncertainty of the aerosol particle number concentration and 10 % of the aerosol particle size measurements, 20 % uncertainty assumed for the impactor loss curve and a 40 % uncertainty due to the aerosol concentrator curve. For the predicted [INP] the ambient size distribution of particles between $0.5 \leq d_{ve} \leq 20\ \mu\text{m}$ was used without further corrections.”

P. 16, L29, conclusions: you mention the good correlation of INP to bulk dust mass, but not the much higher correlation of $R=0.95$ to the total particle number N_{tot} (Fig. 9a), why ?

We mentioned only the bulk dust mass here with the reasoning to make a fair comparison to the other chemical elements, since the dust and other chemical elements mass was derived from the filter measurements while the particle number from other instruments (SMPS, APS, WIBS). However, we have added now also the correlation with the particle concentration of particles larger than $0.5\ \mu\text{m}$. We do not use N_{tot} from Table 3 ($R = 0.95$) because this only refers to particles larger than $0.8\ \mu\text{m}$ (as measured with the WIBS) but instead use the one from Fig. 5 ($R^2 > 0.75$), referring to particles of $d_p \geq 0.5\ \mu\text{m}$ (from SMPS+APS):

P. 17, L. 17-18 now reads: "Submicron INP concentrations in the condensation mode at 240 K were observed to correlate well with the concentration of particles larger than 0.5 μm ($R^2 > 0.75$). Furthermore, they correlated fairly well with the bulk dust mass of particles smaller ..."

P.17, L.5-8, conclusions: you could make a more forceful point of your finding, that ammonium sulfate at the surface of dust increases nucleating properties, by comparing it to the traditional wisdom that insolubility is required for an INP. Pruppacher and Klett (1980) have a whole little chapter 9.2.3.1 named "Insolubility requirement" on that.

We have added the following sentence on P.17, L. 29-31:

"The observation in this work that the presence of a soluble salt ion leads to an improved ice nucleation ability of dust particles questions the conventional assumption of insolubility as a requirement for INPs (Pruppacher and Klett, 1997)."

Technical corrections

P.11, L.34: Fig. 6e must be 7e.

This has been changed (P.12, L12 revised manuscript. "8e" now because of the new Figure 1).

Fig. 10, caption, last sentence: change to "Each ambient PSD curve was measured ...", because the red PINC PSD curves were calculated, as stated in chapt. 3.7.

The sentence has been changed to (now caption Fig.11, P.36):

"Each ambient PSD curve was measured at noon of the respective day."

P.15, L.19/20 and Fig. 12: Fi. 12 has red symbols, which are described neither in the text nor in captions of Fig. 12 or Fig.2. What is it?

We have replaced the sentence in the caption of Fig. 12:

"Color coding is as in Fig. 2 with CLACE2014 data shown in blue. "

to (now Fig.13, P. 38):

"Green data points refer to biomass burning events, orange and red points to intermediate and major dust events, respectively, and black data points to the remaining time periods. CLACE2014 data are shown in blue."

Fig. 5: the dashed vertical lines indicating water saturation do appear in my printout only for 240 K, but not for 233K and 248K. Maybe the whole Figure can be enlarged ?

We have enlarged the figure (now P.31) and made the vertical lines thicker. We will keep it in mind for the final version of the paper.