

We thank Reviewer 2 for their constructive comments. We reproduce reviewer comments in *blue* in the following.

*Page 1, Line 14-19: Can you summarize the impact of aging on the deposition nucleation and the condensation nucleation separately? Also, I think it is justified to say the INP measurements and analysis suggest the aging process in SAL can lead to an increase ice nucleation efficiency of Sahara mineral dust, but in my opinion the overestimation of INPs by D10 and D15 (using the observed aerosol properties) does not deliver the same message. Many data used to derive D10 and D15 parameterizations were collected over the Pacific and western/central US, which are far from the Sahara and are more affected by East Asian dust and local dust sources. If the authors indeed want to convey this message (as the current text shows), additional evidences are needed.*

We do not intend to suggest that aging processes in the SAL lead to enhanced ice nucleation ability solely by comparing to the D10 and D15 parameterization. We have made changes to the abstract and the conclusion section as listed below to stress that we base the conclusion about the enhancing effect of aging first of all on our observations regarding ammonium sulfate and the biological particles. We acknowledge that the comparison to the D10 and D15 parameterizations is not evidence of aging increasing IN ability but rather alludes to the same point. Particularly the comparison with D15 is valuable in this respect because D15 is based on dust from Asia and the Sahara.

We have replaced P.1, L.14-15:

*“We find that an increase of ammonium sulfate, linked to anthropogenic emissions in upwind distant anthropogenic sources, mixed with the desert dust, has a small positive effect on the INP per dust mass ratio. Furthermore, the relative abundance of biological particles was found to be significantly higher in INPs compared to the ambient aerosol. Two common parameterization schemes for INP concentrations, which were derived mostly from atmospheric measurements far away from the Sahara, were found to predict more INPs based on the aerosol load than we observed in the SAL. Overall, this suggests that atmospheric aging processes in the SAL can lead to an increase in ice nucleation efficiency of mineral dust from the Sahara.”*

with P.1, L.14-21 (new manuscript):

*“We find that an increase of ammonium sulfate, linked to anthropogenic emissions in upwind distant anthropogenic sources, mixed with the desert dust, has a small positive effect on the condensation mode INP per dust mass ratio but no effect on the deposition mode INP. Furthermore, the relative abundance of biological particles was found to be significantly higher in INPs compared to the ambient aerosol. Overall, this suggests that atmospheric aging processes in the SAL can lead to an increase in ice nucleation ability of mineral dust from the Sahara. INP concentrations predicted with two common parameterization schemes, which were derived mostly from atmospheric measurements far away from the Sahara, but influenced by Asian and Saharan dust, were found to be higher based on the aerosol load than we observed in the SAL, further suggesting aging effects of INPs in the SAL.”*

Furthermore we have changed the sentence on P.17, L.13:

*“This could be an indication that atmospheric processing as it occurs during transatlantic or transeuropean advection of dust may enhance the ice nucleation ability of mineral dust compared to that after a relatively short atmospheric transport between the Sahara and Tenerife.”*

to (new manuscript) P.18, L.3-7:

*“The enhancing effect of ammonium sulfate on ice nucleation, the higher number of FBAPs in INPs compared to the total ambient aerosol and the comparison particularly to the D15 parameterization could be an indication that atmospheric processing as it occurs during transatlantic or transeuropean advection of dust may enhance the ice nucleation ability of mineral dust compared to that after a relatively short atmospheric transport time between the Sahara and Tenerife.”*

*Page 3, Line 13: It should be noted that while Sullivan et al. (2010) shows nitric acid can lead to higher ice nucleation rate under supersaturated conditions, it also inhibits the deposition nucleation (sub-saturated).*

We have changed P.3, L.11-14:

*“Condensation of sulfuric acid (Knopf and Koop, 2006; Sihvonen et al., 2014; Wex et al., 2014) was observed to mostly impair ice nucleation, whereas ammonium (Salam et al., 2007; Koop and Zobrist, 2009), nitric acid (Sullivan et al., 2010), or the exposure to ozone (Kanji et al., 2013) can promote it.”*

to P.3, L.11-15:

*“Condensation of sulfuric acid (Knopf and Koop, 2006; Sihvonen et al., 2014; Wex et al., 2014) was observed to mostly impair ice nucleation, whereas ammonium (Salam et al., 2007; Koop and Zobrist, 2009), or the exposure to ozone (Kanji et al., 2013) can promote it. Sullivan et al (2010) observed that nitric acid promoted ice nucleation above water saturation but inhibited deposition nucleation below water saturation.”*

*Page 4, Line 22: Could you please elaborate why  $RH_w = 92\%$  and  $RH_w = 105\%$  were chosen for the measurement setup? If a small perturbation was added to it, would the result be sensitive to the change?*

We have added the following sentence on P.4, L.23-26 in the revised manuscript to explain the choice of conditions:

*“These conditions were chosen such that a high enough fraction of the dust particles should activate as INP to be measurable with PINC, to be able to clearly distinguish between deposition and condensation/immersion mode and to compare to an earlier study on free tropospheric INP at the Jungfraujoch in the Swiss Alps conducted under similar conditions.”*

*Page 5, Line 3: Is the size threshold (>3 micrometer) the only criteria to distinguish ice crystals from droplets? What is the typical size of the droplets measured in PINC?*

The size of the droplets depends on the  $RH_w$  conditions in the chamber as well as the temperature. Indeed under certain conditions the droplets can grow to 3  $\mu\text{m}$  as well. We have added now a part on

the evaporation section of PINC which describes in more detail why the size threshold of 3  $\mu\text{m}$  can be used as sole criterion to distinguish ice crystals from droplets and interstitial aerosol.

We have replaced P.4, L.32 – P5., L.4 in the original manuscript:

*“An impactor with an aerodynamic  $D_{50}$  cut-off diameter of 0.9  $\mu\text{m}$  (diameter at which 50 % of the particles impact) was used upstream of PINC to allow a distinction by size of larger ice crystals which had formed in PINC and unactivated aerosol particles and droplets. Ice crystals, droplets and aerosol particles in the size range 0.5-25  $\mu\text{m}$  were detected with an Optical Particle Counter (OPC; Lighthouse REMOTE 5104; Fremont, USA) downstream of PINC. Particles larger than 3  $\mu\text{m}$  were classified as ice crystals.”*

with P.5, L.3 – 11 in the revised manuscript:

*“In the standard set-up an impactor with an aerodynamic  $D_{50}$  cut-off diameter of 0.9  $\mu\text{m}$  (diameter at which 50 % of the particles impact) was used upstream of PINC to allow a distinction by size between larger ice crystals which formed in PINC and unactivated aerosol particles and droplets. In the evaporation section at the lower part of PINC the wall temperatures are kept both at the warm wall's temperature, maintaining  $RH_i = 100\%$  while  $RH_w < 100\%$ , leading to droplet evaporation while ice crystals are preserved. Ice crystals, droplets and aerosol particles in the size range 0.5-25  $\mu\text{m}$  are detected with an Optical Particle Counter (OPC; Lighthouse REMOTE 5104; Fremont, USA) downstream of PINC. Particles larger than 3  $\mu\text{m}$  are classified as ice crystals. Under high  $RH_w$  conditions, droplets may grow to sizes larger than 3  $\mu\text{m}$  and a differentiation by size is not possible anymore. This droplet breakthrough occurs at  $RH_w = 108\%$  for our sampling conditions ( $T = 240\text{ K}$ ).”*

*Page 7, Line 19: Do you mean “analysis”? Reanalysis data are often at coarser resolutions.*

Yes, this was wrong. We have corrected it to (P.7, L.25-26 in the revised manuscript):

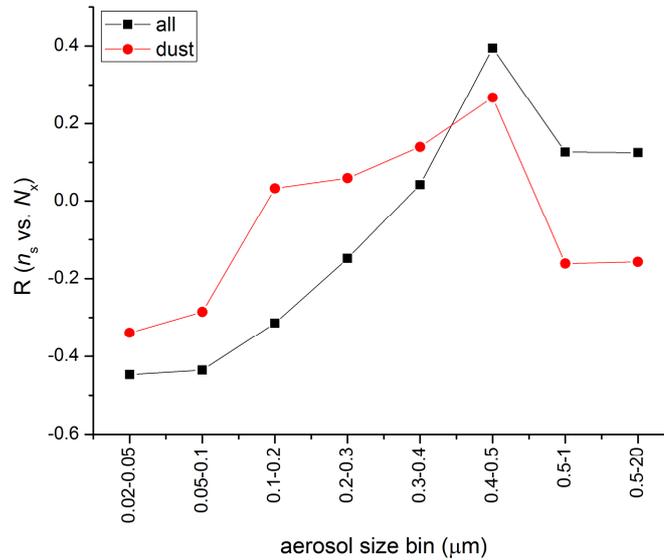
*“ECMWF analysis data were used as input and the model was run with a resolution of 0.25°.”*

*Page 10, section 3.3: The analysis and discussion here are very interesting and useful. Would it be useful to calculate the  $n_s$  function for smaller particles (< 0.5 micro m.) and larger particles separately and compare them?*

Unfortunately, we cannot allocate the fractions of INPs smaller than 0.5  $\mu\text{m}$  and larger than 0.5  $\mu\text{m}$ . Thus, it is impossible to calculate  $n_s$  for the two different size fractions independently. To answer the reviewers comment to see if there is a relation between  $n_s$  and the small particle fraction, we have calculated  $n_s$  for the full size range and made the same comparison with the number of aerosol particles in each size bin as we had done in the manuscript with the INP (see below). Please note that we have plotted R instead of  $R^2$  here.

The correlation of the INP concentrations with the size bins (as done in the manuscript) warrants the usage of  $n_s$  since larger size bins correlate better with the INP concentrations. Correlating now  $n_s$  with particles in the different size bins, the clear dependency of INP on size gets lost. This is because  $n_s \approx \text{INP}/(\pi d^2)$ , hence the denominator and numerator are proportional to  $d^x$ . We don't know the exact dependency of INP on  $d$  but expect  $x$  in this case to be close to 2 since ice nucleation is a surface process.

We see now in the plot below that R peaks for aerosol particles of  $d_p = 0.4-0.5 \mu\text{m}$ , which are the particles providing the highest fraction to the total surface area during most of the CALIMA campaigns. However, all R values are very low. Since we don't think the plot is helping the reader to better understand the size dependency of ice nucleation, we do not include this plot in the revised manuscript.



*Page 13, Fig2: Is there a particular reason for using “.8” on the time axis? Would be nice to use integer numbers. Is the time local time?*

“.8” was referring to August and was missing the “.” after “8”. We've changed the date format now from “DD.M” to “DD.MM.” in Fig. 3, Fig. 4, Fig. 7 and Fig. 12 of the revised manuscript.

*Page 14, Line 15: Does this limitation also apply to other types of instruments? In other words, is the poor relationship between INP and the number of >0.5um particles solely because of the instrument imitation? Please comment on this.*

This limitation is common for CFDC-like instruments which use size as criterion for differentiation between droplets and ice crystals. The impactor's  $D_{50}$  of different instruments varies and the one of PINC is rather at the lower end. This is why we provide this discussion. However, the correlation during the CALIMA campaigns between the condensation INP concentrations at 240 K and aerosol particles  $> 0.5\mu\text{m}$  is not poor ( $R^2 > 0.78$ ). It only is so for the deposition mode INP at 240 K ( $R^2 < 0.2$ ). The poor correlation with the deposition mode INP likely is due to the low activated fractions at this  $RH_w$  which is more than a factor 15 lower than that of the condensation mode INP.

*Page 29, Fig5: This figure is very informative. I think it is important to mention that the derived ns functions can differ at about one magnitude between various dust events.*

We fully agree. We have stated on P.11, L. 1 (original manuscript):

*“In addition, differences between the different SDEs can be found by up to an order of magnitude which must be related to the composition of the aerosol particles.”*

and have replaced it with P.11, L.12-13 (new manuscript):

*“In addition, differences of up to one order of magnitude in  $n_s$  between the different SDEs are found which must be related to the composition of the aerosol particles.”*

*Page 36, Fig12 caption, Line 2: “Color coding is as in Fig 2...” What does black color indicate? Background conditions? Please consider adding a legend for convenience of the reader.*

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 caption of 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.”*

*Page 36, Fig12 caption: It would be useful to provide a formula showing how the uncertainties were calculated/combined.*

The uncertainties have been combined by standard error propagation. However, we realized that we have not yet provided the uncertainties we took into account. This has been added now, in addition to adding an equation on how the INP concentrations including the omitted INPs have been calculated.

Equation 2 on the omitted INPs has been added on P.15, L.9:

$$INP_{\text{omitted}} = n_s A_{\text{omitted}}$$

We have added on P.16, L.8-10 (revised manuscript):

*“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.”*