Anonymous Referee #1

For clarity, the referee's comments are copied in black and our responses are offset in blue.

Summary: The manuscript by Crawford et al. (2015) presents results from 12 days of fluorescent aerosol measurement during winter time at the Jungfraujoch, Switzerland, an observatory at 3580 m altitude. Measurements were conducted with the wide-band integrated bioaerosol spectrometer (WIBS-4). A recently introduced cluster algorithm (Crawford et al. 2015) was applied for the statistical analysis of fluorescent particles. The analysis revealed that almost all fluorescent particles measured were mineral dust and only a minority of biological origin. Based on the low number concentration of primary biological aerosol particles (PBAP) observed in this study, a maximum ice active fraction of 0.5% at -9.7C reported by Mohler et al. (2008) for a common bacterial strain, Pseudomonas syringae, and the several order of magnitude larger ice crystal concentrations observed at Jungfraujoch, it is concluded that PBAP do not significantly contribute to ice crystal concentrations at this site during winter time.

The paper is significant in that there are currently only few observations of biological aerosol particles and cloud interactions during winter time and it represents an additional application of the new clustering algorithm introduced by Crawford et al. (2015). However, the current manuscript shows several deficiencies. The discussion of the observations, their uncertainty and shortcoming, and the implication of the results are often kept at a minimum. There are several incidents were related work is not cited sufficiently and assumptions being made without discussion of their validity. The general structure of the manuscript is good, however, long sentences make it hard for the reader to follow. Overall, the manuscript gives the impression that the authors did not invest much effort in preparing it. This is a pity because the measurements and results themselves would certainly be of interest to the readers of ACP.

Therefore, I only suggest publication of the manuscript in ACP if major revisions are undertaken and the following remarks are taken into consideration.

We thank the reviewer for their helpful comments and recommendations which we address below.

General remarks:

The title stresses that the measurements were conducted and are representative for free tropospheric conditions. However, the manuscript completely lacks a confirmation and discussion of this truly being the case. The rather old publication (Baltensperger et al. 1998) which is given as reference already states that "during winter the site represents the free troposphere most of the time", but not all the time, as the current manuscript suggests. A recent study by Herrmann et al. (2015) showed that this is the case "over 60 % in January". I advise the authors to be more careful with the claim of measuring in the free troposphere and investigate if this is applicable for their measurement period.

We thank the reviewer for their useful suggestion. We now include a discussion of free tropospheric conditions in section 3.1. In this discussion we use the concentration of particles larger than 90 nm in diameter (N_{90}) as described in Herrmann et al. (2015) to distinguish periods of free tropospheric conditions from those influenced by planetary boundary layer (PBL). They found that N_{90} = 40 cm^{-3} was a good approximation to describe free tropospheric background aerosol across all seasons, with periods influenced by the PBL resulting in N_{90} concentrations of several hundred to 1000 cm^{-3}. These values were found to be lower in winter so we use $N_{90} < 30$ cm^{-3} to be representative of background FT conditions and $N_{90} < 50$ cm^{-3} to be representative of "FT-like" conditions during the sampling period as described in Herrmann et al. (2015).

A time series of N_{90} concentration for the period used in this manuscript is presented in Figure 1 where the background FT condition of $N_{90} < 30$ cm^{-3} is met 66.2% of the time and "FT-like" conditions where $N_{90} < 50$ cm^{-3} is met 88.4% of the time and we use this higher limit to define FT-like conditions in our analysis. Periods with $N_{90} > 50$ cm^{-3}, such as the extended period between 09:00 15/02 – 09:00 16/02, are excluded. This figure will be included in the revised manuscript along with a short discussion of the FT conditions during the sampling period in section 3.1.

This analysis was performed by Erik Herrmann and Christopher Hoyle at the Paul Scherrer Institute and they are added as co-authors in the revised manuscript for their contribution.

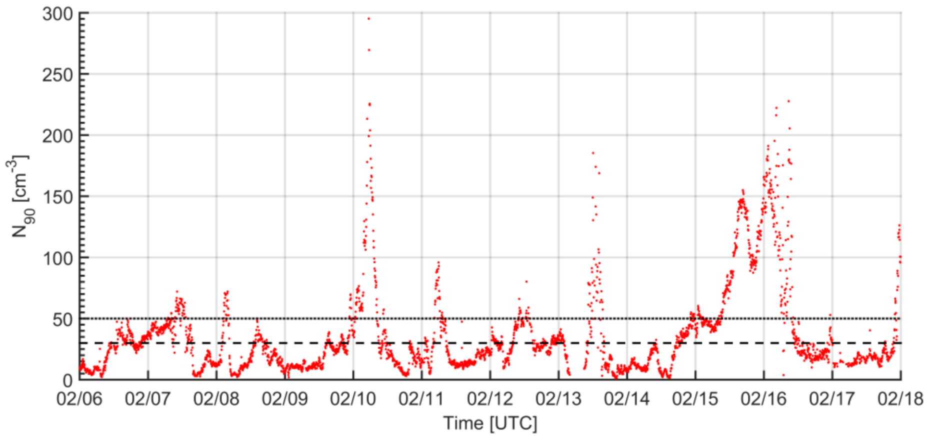

Figure 1. Time series of SMPS N_{90} concentration for the analysis period. Dashed line denotes the 30 cm^{-3} background concentration described in Herrmann et al. (2015); the dotted line denotes this 50 cm^{-3} theshold used to distinguish free tropospheric conditions.

The manuscript claims to have measured a representative time period of the typical background aerosol concentrations at the Jungfraujoch during wintertime. Which indicators have been used to support this claim? Has a comparison been done to long term measurements at the same site during winter time using other instrumentation? Considering the short measurement period of only 12 days and the rather uniform origin of air masses from over the Atlantic ocean, as mentioned in the manuscript, the representativeness of the measurements for "typical background aerosol concentrations" are questionable. Supportive data needs to be presented in the manuscript.

We will include a comparison of aerosol data collected during the campaign to long term measurements made at the site in the revised manuscript in section 3.2; Figure 2 shows median, 25th percentile and 75th percentile SMPS and OPC size resolved concentration measurements made during the month of February from 2009 to 2014 which we compare to the campaign median SMPS, OPC and WIBS NonFl and Fl size resolved concentrations. It can be seen that the campaign measurements typically lie within the range of the 25th percentile and median values of the long term measurements during February at the site, suggesting that the measurement period can be considered to be representative of the typical FT background aerosol concentration at the Jungfraujoch during wintertime.

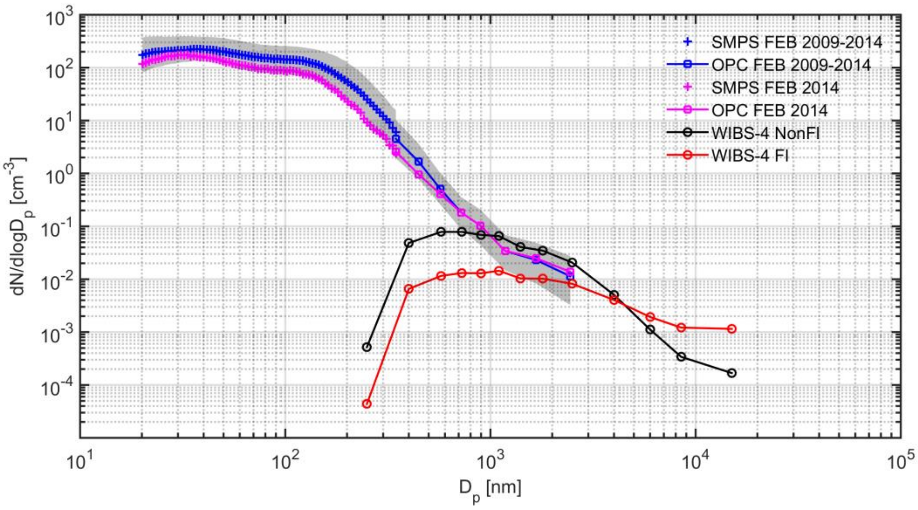

Figure 2. Comparison of long term median SMPS and OPC size resolved concentration measurements made during February 2009 to 2014 to those made during the 2014 campaign. Grey shaded area represents the 25th and 75th percentiles of the long term measurements.

How does the statement in the introduction "even modest concentrations of primary ice can result in the rapid glaciation and subsequently cause precipitation: : :" and the conclusions at the end of Section 4, "such low concentrations of PBAP are unlikely to have any significant impact on cloud evolution through ice nucleation (: : :) IN concentrations of only 5x10-4 L-1"? This can only go together if you clearly define "modest" and give typical number concentrations of ice nuclei found to impact cloud evolution.

The statement in the introduction refers to cases where secondary ice production via the Hallet-Mossop (HM) process has caused rapid glaciation in clouds which contained low concentrations of primary ice. In this study the majority of cloud events occurred outside of the Hallet-Mossop zone, thus secondary ice production via the HM process as discussed in Lloyd et al., (2015) companion study. We will clarify this in the revised manuscript and we will include a discussion on the possibility of secondary ice production via the HM process in section 4.

In the part about the cluster analysis and its interpretation it is almost impossible for the reader to follow as the cluster algorithm is not described nor are details given about the interpretation of the fluorescence analysis. What are physical differences between particles in cluster 1 and 2? How likely is it that cluster 3 is representative for Pseudomonas syringae?

A sufficient summary of the methodology used is given at the start of section 4 and a full description of why this methodology was chosen is presented in Crawford et al., (2015) which is cited on pg 26075, ln 7. We see no reason to repeat the rationale presented in Crawford et al., (2015) here.

The key physical differences between clusters 1 and 2 are that cluster 1 is much larger and more aspherical than cluster 2.

Without supporting measurements we cannot identify the origin of cluster 3 beyond suggesting that it is likely biological, given its large size, asymmetry and moderate fluorescence. We use *Pseudomonas syringae* as an illustrative example in the discussion of how this cluster may act as source of ice via primary ice nucleation as the ice activity of this species has been well characterised in laboratory experiments under atmospherically relevant conditions. We do not wish to suggest that this cluster is representative of

Pseudomonas syringae, we are simply using this assignment as a discussion point for the clusters potential impact on cloud microphysics. We agree to clarify this in the revised manuscript.

A general technical comment: It is not specified if the presented concentrations are given at local conditions or if they have been normalized to standard temperature and pressure conditions. The latter would be recommended. Please clarify.

Concentrations are given at local conditions in keeping with previous reports from this site, e.g., Herrmann et al., (2015).

Specific remarks:

p 26068 l25: define "modest", otherwise this appears as a contradiction to your own results

Crawford et al., (2012) showed that low concentrations of primary ice (~ 0.01 L^{-1}) resulted in the rapid glaciation of a shallow convective wintertime cumulus via the Hallet-Mossop ice multiplication process. In this study secondary ice production via the Hallet–Mossop process was ruled out as the clouds observed were rarely within the active temperature range for this process as discussed in Lloyd et al., (2015) companion study; Glaciation via the Wegener-Bergeron-Findeisen process was ruled out as the observed updraft velocity exceeded the minimum threshold required for the co-existence of liquid water and ice crystals in mixed phase cloud for the majority of the campaign as discussed in the Farrington et al., (2015) companion study. As such we concluded that biological IN were not significant at the site during the measurement period. We will clarify these points in section 1 and 4 of the revised manuscript.

P26069 l8ff: What kind of coatings are you referring to? Not all coating necessarily increase the saturation ratio required for ice nucleation. Please provide citations to studies you are referring to

Here we are referring to secondary organic aerosol (SOA), sulphuric acid and ammonium sulphate coatings; Möhler et al., (2008) and Koehler et al., (2010) showed that Arizona test dust (ATD) coated with SOA significantly increases the critical ice saturation ratio for nucleation compared to untreated ATD; Similarly sulphuric acid and ammonium sulphate coatings have been found to generally act to increase the critical ice saturation ratio for nucleation compared to untreated mineral dust (Cziczo et al., 2009; Eastwood et al., 2008; Chernoff & Bertram 2010; Sullivan et al., 2010). We will refer to these studies in the revised manuscript.

P26070 l6: reference to some of these campaigns?

We will include references to these campaigns in the revised manuscript.

P26071 l8-26: References for the description of the WIBS-4 are completely missing in the paragraph. Please cite them appropriately.

We agree to revise this section to include additional references.

P26072 l11: Please give a brief summary of the agglomerative data processing method you are using in the current manuscript. The reader should be able to understand and follow your method without reading another paper.

This is described later in the manuscript. We will make the following change in the revised manuscript:

"In this study we use a new hierarchical agglomerative data processing method for WIBS-4 UV-LIF measurements to discriminate between particle types and methods used are described in section 4."

P26073 l16: A description of the surrounding of the Jungfraujoch is necessary for readers not being familiar with the local terrain (e.g. Aletsch glacier). Even a topographical map could be added.

We thank the reviewer for their helpful suggestion and we will include a description of the site surroundings in section 2.1.

Figure 3: Indicate the in cloud and out of cloud periods in this figure since you are referring to it when talking about average in cloud and out of cloud Nfl and Ntot

We thank the reviewer for their helpful suggestion and we will include a shaded area in the figure to indicate in-cloud periods.

P26074 l4ff: Which role does the total inlet play here? Were differences expected between in cloud and out of cloud cases? Which implication does the observed temperature dependence of the fluorescent aerosol fraction have? Please discuss your results more.

The results in this section have changed due to filtering out PBL influenced air masses in the revised analysis and increasing the ice mass fraction used to define the threshold between mixed phase and glaciated condition from IMF ≥ 0.5 to IMF ≥ 0.9. These changes are discussed in the response to referee #2 and will be discussed in detail in the revised manuscript.

The total inlet is used throughout and samples all particles with D_p < 40 µm where the sample air is heated to evaporate droplets and ice crystals such that their residuals are sampled along with the interstitial aerosol.

Figure 7: uncertainty bands? Since at large sizes only very few particles are counted, the uncertainty must be much larger than at the small sizes I suspect?

For clarity and ease of comparison of the different cases we only show the averages in this figure but we agree to include individual plots for each case showing the standard deviations in an appendix which is shown in Figure 3.

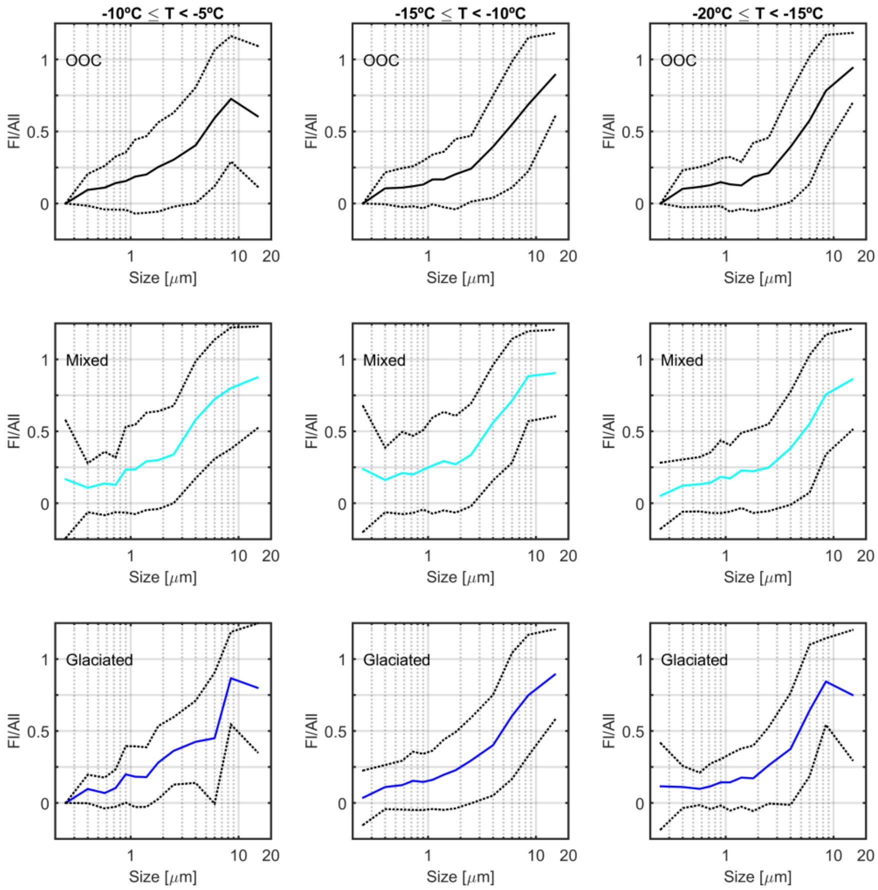

Figure 3. Size dependent fluorescent aerosol fractions for out of cloud (black, top row), mixed phase (cyan, middle row) and glaciated conditions (blue, bottom row) over the three different temperature regimes studied (columns).

P26074 l16: which meteorological and cloud microphysical parameters have you investigated? Please specify. Have you only looked at these time series or done correlation and more in depth analysis of trends?

We have investigated the trends and correlations between mean and median fluorescent aerosol fraction and the following meteorological and cloud microphysical parameters; ice mass fraction (IMF); total water content (TWC); ice water content (IWC); liquid water content (LWC); ice and droplet concentrations; temperature; wind speed and direction. A scatter plot of the mean (black +) and median (red diamonds) values for each cloud event is shown in Figure 4, along with the corresponding r^2 value where no significant correlation between parameters is observed. We will include this figure in the revised manuscript along with a discussion of the results in section 3.2.

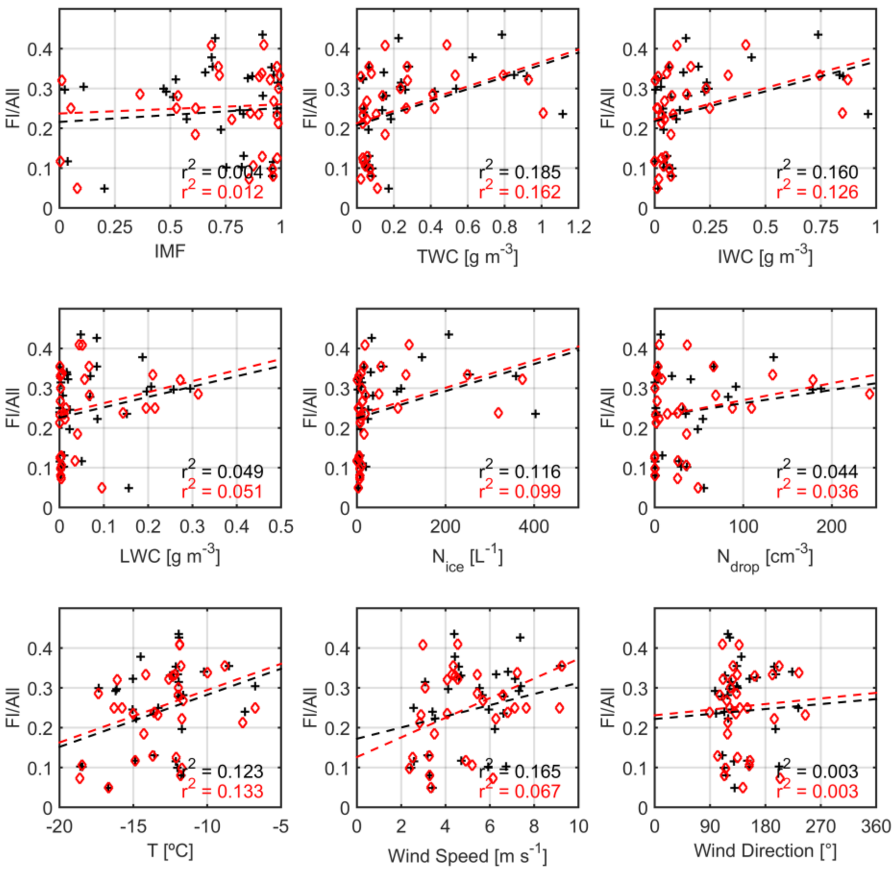

Figure 4. Correlation scatter plot of the fluorescent aerosol fraction to ice mass fraction (IMF); total water content (TWC); ice water content (IWC); liquid water content (LWC); ice crystal and droplet number concentrations; temperature; wind speed and direction for cloud events persisting for at least 30 min in duration. Mean values are denoted by black + symbols and median values by red diamonds.

P26075 l12-16 It's impossible to compare the clusters since only for cluster 3 number concentrations are given. The correlation with Nfl of cl1 and cl2 shows that most of the fluorescent particles were found in these two clusters, however, a simple number concentration provide more insight.

> We will include the campaign average concentrations for clusters 1 and 2 in the revised manuscript.

P26075 l18: This is unclear. "lower" than what? Are you saying you expected lower concentrations than you measured or what you measured is what you expected?

> We will revise this to:
>
> "We would expect low concentrations of local PBAP in the wintertime…"

p26075 l21: This goes back to the major comment about the free troposphere claim: if you have any planetary boundary layer influence at all, pure free tropospheric conditions are not given.

As discussed in an earlier response we use the concentration of particles larger than 90 nm in diameter (N_{90}) as described in Herrmann et al. (2015) to distinguish periods of free tropospheric conditions from those influenced by planetary boundary layer using a conservative threshold of $N_{90} < 50\ cm^{-3}$ to reject PBL influenced air.

P26075 l27: there are more measurements of biological ice nuclei available than Mohler et al. 2008. Please also consider them

We use the ice active fractions reported for *Pseudomonas syringae* in the Möhler et al., (2008) study for illustration here as the characterisation was performed under atmospherically relevant conditions using the AIDA aerosol and cloud simulation chamber. The majority of experiments studying biological particles use cold stage droplet-freezing assays (e.g., methods used in Vali, 1971; Vali et al., 1976) such as the recent Morris et al,. (2013) study which demonstrated fungal rusts forming ice at temperatures greater than -10°C. While these approaches are useful for identifying ice active particles, caution must be taken when deriving ice activation efficiencies using these methods as significant discrepancies between cold stage wet-suspension methods and dry-dispersion cloud chamber simulations have recently been demonstrated at warm temperatures (Emersic et al., 2015). However, in the case presented here even if cluster 3 was 100% ice active it would still only contribute negligibly to the observed ice concentration.

p26076 l7-12: Be careful with such general statements. Your measurement period was very short and if at all can be representative for winter time. This should be clarified here.

We will revise this to:

"we report that there was no apparent link between the fluorescent aerosol fraction and observed cloud microphysical parameters and meteorology, suggesting that aerosol fluorescence did not influence cloud formation/evolution at the site during the measurement period."

Technical remarks:

All figures should be made bigger and the font size needs to be larger. The axis labels are just at the edge to be readable.

We will increase the figure and font size in the revised manuscript.

The official name of the Jungfraujoch observatory is "High Altitude Research Station Jungfraujoch": please correct this throughout the paper, especially in the title

We thank the reviewer for the correction and we will apply this throughout the revised manuscript.

P26069 l19: be consistent with the spelling of "Primary Biological Aerosol Particles": in the abstract it is spelled with lower case

We will ensure that this is consistent in the revised manuscript.

P26069 l19: insert "(PBAP)" after "Primary Biological Aerosol Particles"

This will be corrected in the revised manuscript.

p26069 l26: the order of citations is not consistent throughout the manuscript. Sort them consistently either chronologically or alphabetically

We will ensure that this is consistent in the revised manuscript.

p26069 l27: Please make at least two sentences out of this very long one

We will make the requested revision.

p26070 l5: replace “Alpine” with “altitude”

We will correct this in the revised manuscript.

p26070 l6: insert “-“ between “cloud” and “aerosol”

We will correct this in the revised manuscript.

p26070 l11: replace “D” with “diameter, Dp”

We will correct this in the revised manuscript.

p26070 l20: replace “Alpine” with “altitude”

We will correct this in the revised manuscript.

p26070 l20: define “a.s.l.”

We will define this in the revised manuscript.

p26071 l6-7: order of citations?

We will ensure that this is consistent in the revised manuscript.

p26071 l12: insert “to” before “determine”

We will correct this in the revised manuscript.

p26071 l17: rephrase the sentence. “bands (: : :) are (: : :) recorded” sounds odd.

We will rephrase this to the following in the revised manuscript:

“The detectors are filtered to measure fluorescence over two detection bands (320–400 and 410–650 nm)”

p26071 l19: replace “2nd” by “second”

We will correct this in the revised manuscript.

p26071 l23: delete “to know”

We will correct this in the revised manuscript.

p26072 l1: delete “,” and insert parentheses around “Gabey et al., 2011”

We will correct this in the revised manuscript.

p26072 l17: delete “measurements of”

We will correct this in the revised manuscript.

p26072 l20: insert ")" after 3V-CPI

We will correct this in the revised manuscript.

p26072 l21f: replace " e.g. Lawson et al. (2015)" with "(e.g. Lawson et al., 2015)"

We will correct this in the revised manuscript.

p26072 l23-24: repetition. Please rephrase the sentence.

We will rephrase this in the revised manuscript.

p26072 l29: replace ";" with "and"

We will correct this in the revised manuscript.

p26073 l1: replace "Saharan dust events" with "SDE's"

We will correct this in the revised manuscript.

Figure 4 and 5: What do the whiskers and horizontal lines denote in the different plots?

5th, 25th, 50th, 75th and 95th percentile. We will include this in the figure captions in the revised manuscript.

Figure 6 and Figure 7: replace the x-axis label "size" with "aerodynamic diameter" if it is the aerodynamic diameter which you are showing.

Reported sizes are optical diameter. We will clarify this in the revised manuscript.

Figure 6: caption and title: be consistent: is it "-15CT < 10C" or "-15C < T10C"? This also refers to p26074 l 18.

We will ensure that this is consistent in the revised manuscript.

Figure 6: what do the different line colors for the mean size distribution show? They can all be black

The colours used for the mean size distributions are used to represent out of cloud, mixed phase and glaciated conditions in keeping with Fig. 4 and Fig. 7.

p26074 l 21 replace "Figure" with "Fig."

We will correct this in the revised manuscript.

p26075 l5 replace "size" with "diameter"

We will correct this in the revised manuscript.

p26075 l5 insert "(AF)" after "asymmetry factors"

We will correct this in the revised manuscript.

p26075 l17 delete " of" after "reaching"

We will correct this in the revised manuscript.

p26075 l18: either split the sentence into two or delete the second part of the sentence as this is rather a repetition of the first part. "for very little in the way" sounds colloquial.

We will delete the second part of the sentence.

p26076 l24: split sentence in two

We will revise this sentence in the revised manuscript.

p26076 l25: insert “emissions from “ after “large”

We will correct this in the revised manuscript.

References

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