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.

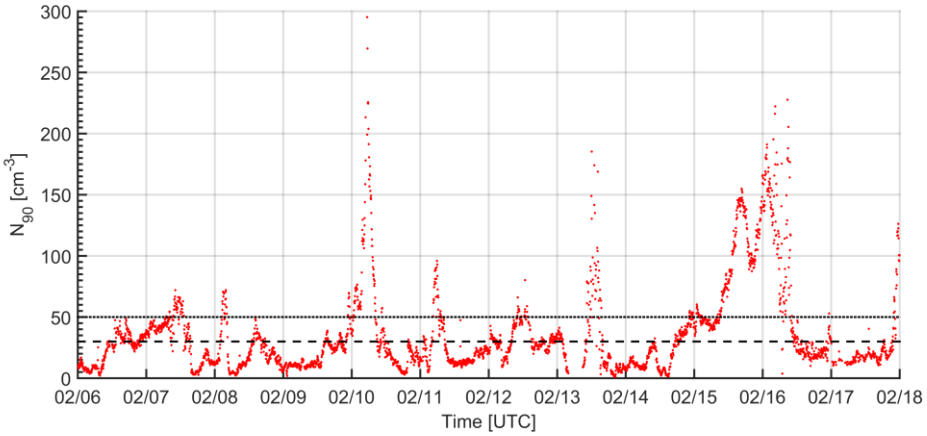

Figure 1. Time series of SMPS N_{90} concentration for the analysis period. Dashed line denotes the 30 cm^{-3} background concentration described in Herrmann et al. (2015); the dotted line denotes this 50 cm^{-3} theshold used to distinguish free tropospheric conditions.

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.

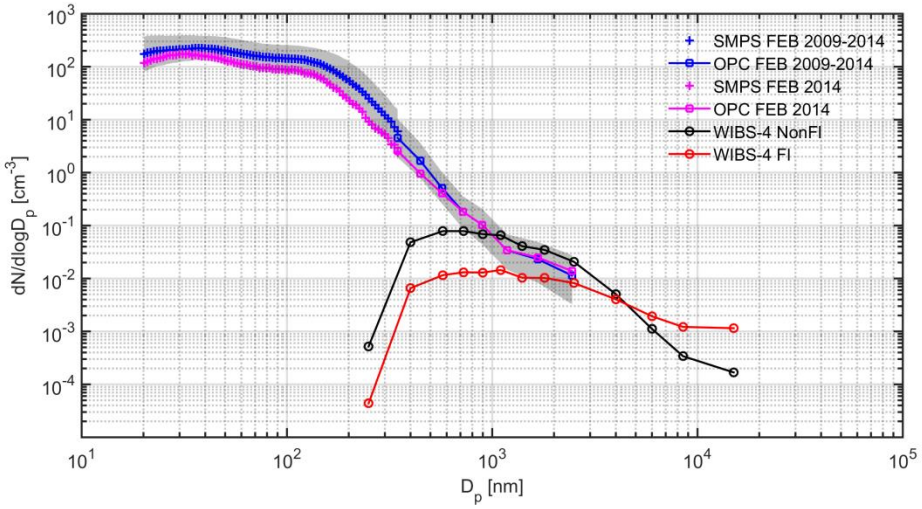

Figure 2. Comparison of long term median SMPS and OPC size resolved concentration measurements made during February 2009 to 2014 to those made during the 2014 campaign. Grey shaded area represents the 25th and 75th percentiles of the long term measurements.

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.

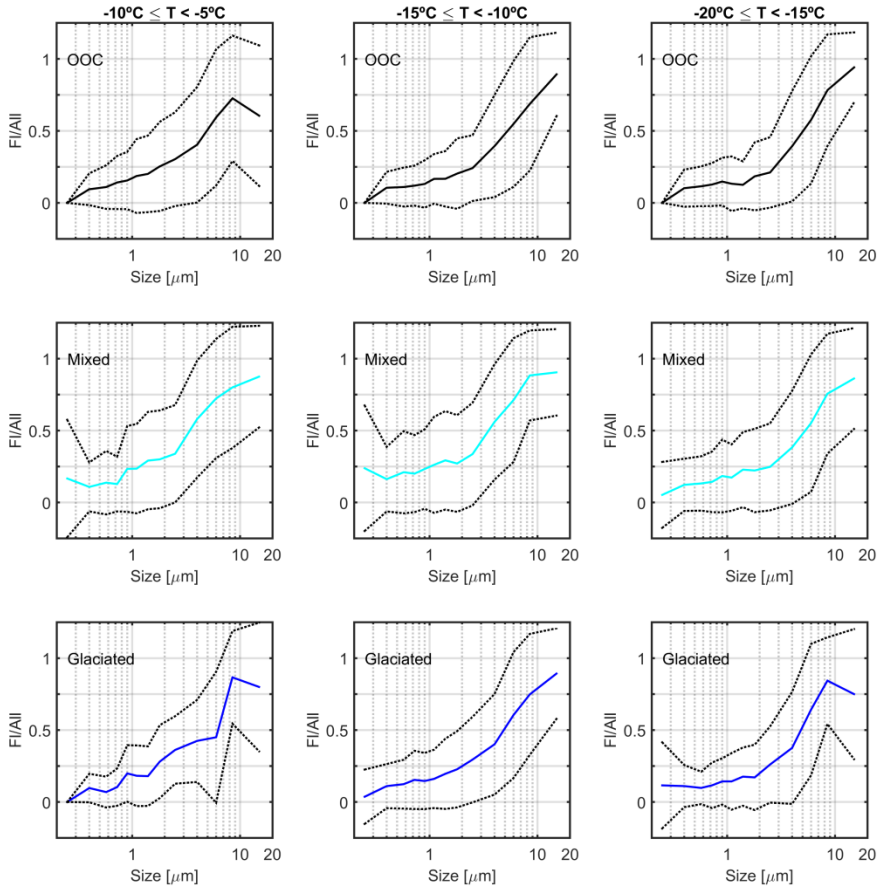

Figure 3. Size dependent fluorescent aerosol fractions for out of cloud (black, top row), mixed phase (cyan, middle row) and glaciated conditions (blue, bottom row) over the three different temperature regimes studied (columns).

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.

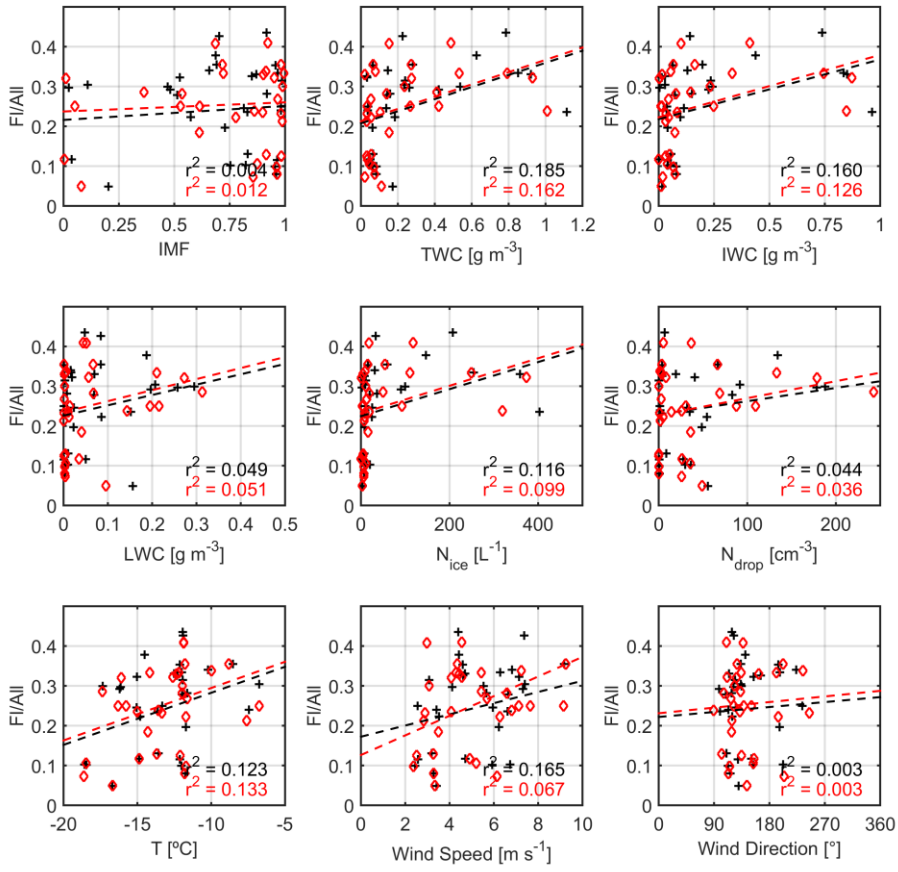

Figure 4. Correlation scatter plot of the fluorescent aerosol fraction to ice mass fraction (IMF); total water content (TWC); ice water content (IWC); liquid water content (LWC); ice crystal and droplet number concentrations; temperature; wind speed and direction for cloud events persisting for at least 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?

5^{th}, 25^{th}, 50^{th}, 75^{th} and 95^{th} 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.

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Anonymous Referee #2

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

General comments: Overall, the work presented in this paper does attempt to address a fundamental question regarding aerosol interactions in the free troposphere and their impact on cloud development. However, I agree with many of the points brought up by Reviewer 1 in that there was a lack of adequate discussion of the results and implications of this work. Additionally, while the data generated in this study is interesting and substantial, there are a few improvements that could be made on the analyses that would help strengthen some of the claims made in the conclusion. I will discuss below the areas that I think could use more attention.

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

Specific comments: As one of the main points of this paper was to investigate the relationship between aerosol particle concentrations and cloud microphysical properties, I suggest that the following be considered and discussed in more detail:

1. Cluster 3 is classified as biological material based on similar fluorescence described in a previous paper (Crawford et. al. 2014). In Crawford et. al. 2014, a more detailed list of airborne bacterial phyla and families as well as a few groups of fungal spores were identified as likely representatives of the fluorescent PBAPs. However, the current paper only discusses the implications of the results under the assumption that cluster 3 represents Pseudomonas syringae (Mohler 2008 and Lloyd 2015). While it is true that the ice-active fraction of P. syringae is low in the environment, is there a possibility that cluster 3 may also represent other ice-active microorganisms found in higher concentrations?

We don't know the origin of the aerosol in cluster 3. We assume it is likely PBAP based on its moderate fluorescence in all three channels and high asymmetry factor but we cannot determine which meta-class it belongs to. We use Pseudomonas syringae for an illustrative example here as it has been well characterised under atmospherically relevant laboratory conditions (Möhler et. al. 2008).

2. The effect of PBAPs on meteorological processes presents an area of research where there are still many uncertainties. As such, the results presented herein on aerosol concentrations in the free troposphere are significant however the implications of the results are only covered briefly, and would benefit from a more detailed discussion. It's concluded that such low concentrations of PBAPs and their estimated ice-active fraction would have negligible influence on cloud properties, with only two papers (Mohler 2008 and Lloyd 2015) referenced. In fact, there exists a body of literature that specifically addresses how similarly low concentrations of INPs may still influence cloud glaciation and precipitation development via secondary ice formation mechanisms (a few of which I have listed below). In particular, I encourage looking through Korolev 2007, which outlines conditions conducive to rapid glaciation of mixedphase clouds through the Wegener-Bergeron-Findeisen mechanism. These conditions may be similar to those of the clouds sampled at Jungfraujoch. These papers also address the discrepancy between ice crystal and ice nuclei concentrations in mixedphase clouds, which is a point used in this current study to back the claim that the fluorescent PBAP concentrations detected are too low to affect nucleation processes (pg. 26076 lines 1-2).

References:

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We thank the reviewer for their helpful suggestions. A thorough analysis of the cloud microphysics data from this experiment is presented in the Lloyd et al., (2015) companion study which concludes that atmospheric secondary ice production contributes negligibly to the observed ice crystal concentration. A second companion study by Farrington et al., (2015) investigated the potential influence of the Wegener-Bergeron-Findeisen (WBF) process at the site where they found that the critical updraft speed (as defined by Korolev and Mazin (2003) and Korolov (2007)) to maintain mixed phase conditions was less than the observed updraft velocity for the majority of the INUPIAQ campaign using the ice 2D-S size distribution as the input for the $N_i r_i$ term. That is why they concluded that glaciation via the WBF process was not significant. Reducing N_i to the projected bio-IN concentrations would reduce this critical threshold significantly, further reducing the influence of the WBF process. A discussion of these processes has now been added to section 4.

3. It is stated that “no apparent trend is observed between mean fluorescent aerosol fractions and contemporaneous mean meteorological or cloud microphysical parameters, suggesting that particle fluorescence does not impact cloud evolution or formation (pg. 26074 lines 14-17),” and again later it is concluded that there is “no apparent link between the fluorescent aerosol fraction and observed cloud microphysical parameters and meteorology, suggesting that aerosol fluorescence does not influence cloud formation/evolution at the site. (pg. 26076 and lines 10-12).” For the reader, it may be difficult to see any trend or lack thereof in this data based solely on figure 5. A statistical analysis on the meteorological/microphysical and fluorescence data (i.e., regression) and including a test statistic and accompanying p-value to back claims that there is no relationship would be helpful.

We thank the reviewer for their helpful suggestions. This is answered in our response to referee 1.

Technical corrections:

Pg. 26068 Line 25: What are “modest” concentrations?

This is answered in our response to referee 1.

Pg 26073 Line. 6: “Discussion of the SDE’s will be described elsewhere.” While you do mention the companion paper to this study in the introduction, it should be clarified here again where there SDE discussions will be taking place.

At the time of writing other participants in the INUPIAQ experiment are in the process of preparing a manuscript investigating the SDE’s using the ice selective inlet.

Pg. 26074 Line 8: What test is used to determine whether there is any statistical significance? Eyeballing standard deviations is not always sufficient for determining significance.

The inclusion of filtering for FT-like conditions as described in our response to referee 1 and the increase of the IMF threshold to differentiate between mixed phase and glaciated conditions from IMF ≥ 0.5 to IMF ≥ 0.9 (see response to Erik Herrmann) to be in line with Lloyd et al., (2015) has produced some significant changes to this analysis and its interpretation. Notably this results in an increase in the mean and median fluorescent fraction for the mixed phase cases compared to the out of cloud and glaciated cases over all temperature regimes as shown in the revised figure below (Figure 1), which now includes the corresponding fluorescent and non-fluorescent aerosol concentrations for comparison.

The observed increase in the fluorescent aerosol fraction in mixed phase conditions is generally a result of a reduction in the non-fluorescent aerosol concentration relative to the corresponding out of cloud cases, rather than an enhancement in the fluorescent aerosol concentration. One possible explanation for this is that non-fluorescent aerosol has been removed via CCN activation and lost in precipitating raindrops in mixed phase clouds as this is not pronounced in the glaciated cases, however, caution must be applied when interpreting the results of this general approach as the differences in fluorescent aerosol fraction may be caused by differences in the sampled air masses for each case.

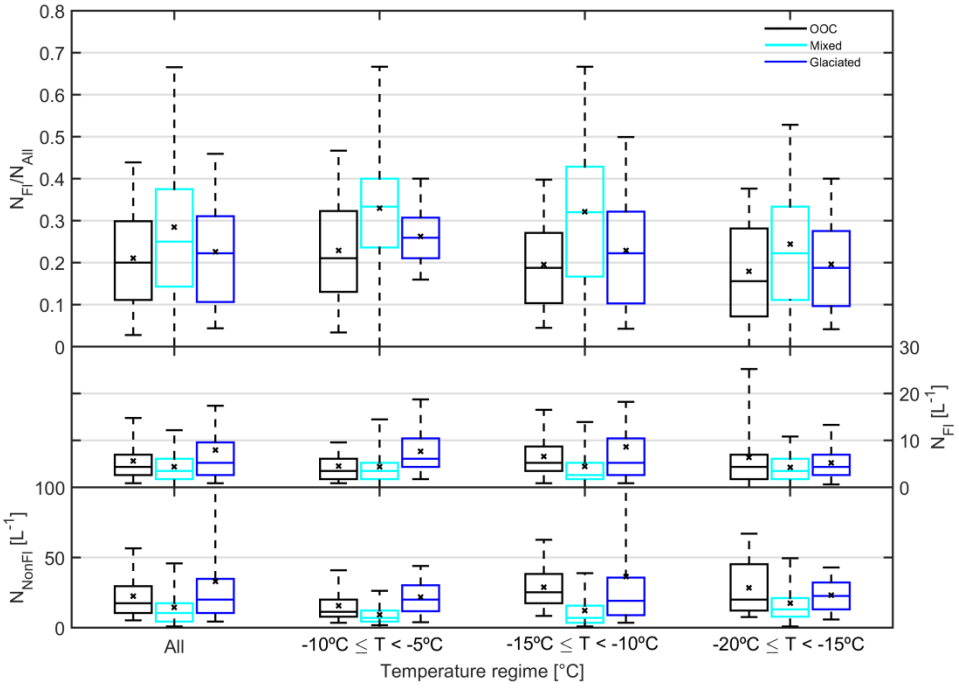

Figure 1. Revised figure 4. Now includes box and whisker plots showing the fluorescent and non-fluorescent aerosol concentrations for each case

We have performed a 1 way ANOVA analysis on the revised data to test for statistical significance which is now described; first we assess the influence of temperature separately for in cloud and out of cloud conditions (TWC ≥ 0.01 gm^{-3}) as shown in Figure 2. It can be seen that in each case the fluorescent fraction decreases with decreasing temperature. The small p-values reported indicate that the means are statistically significantly different; however, the spread in values are large.

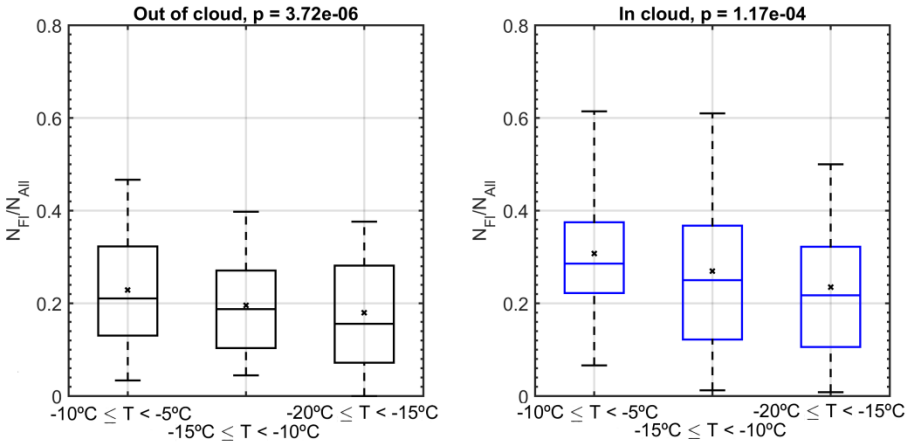

Figure 2. Influence of temperature on fluorescent fraction for out of cloud and in cloud cases.

Next we assess the influence of the presence of cloud on fluorescent fraction at each temperature by comparing the out of cloud and in cloud cases as shown in Figure 3. This shows that fluorescent fraction is increased in cloud.

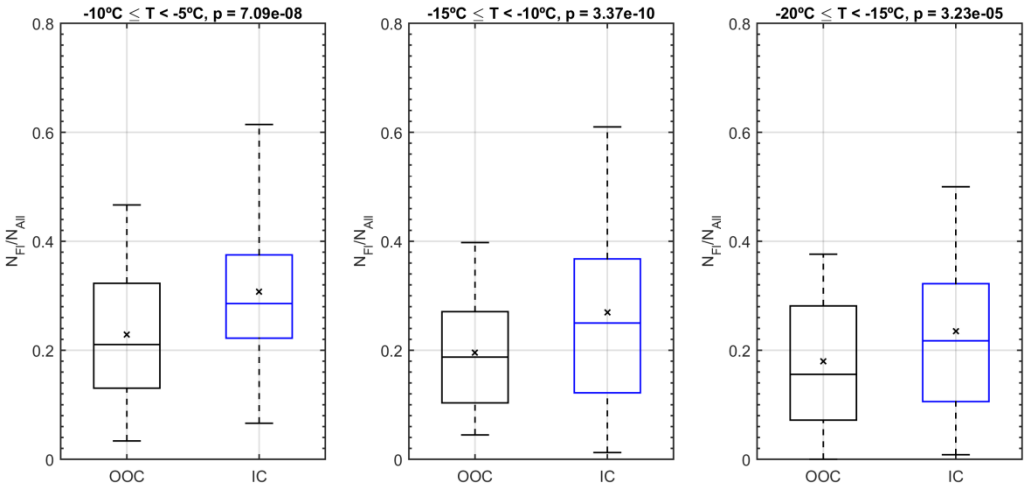

Figure 3. Influence of cloud on fluorescent fraction for the studied temperature regimes.

Finally we assess the influence of cloud type on the fluorescent fraction for each temperature regime as shown in Figure 4. Here, it can be seen that the fluorescent fractions are generally greater in mixed phase conditions than in glaciated conditions.

We will include a discussion of the revised analysis in the revised manuscript.

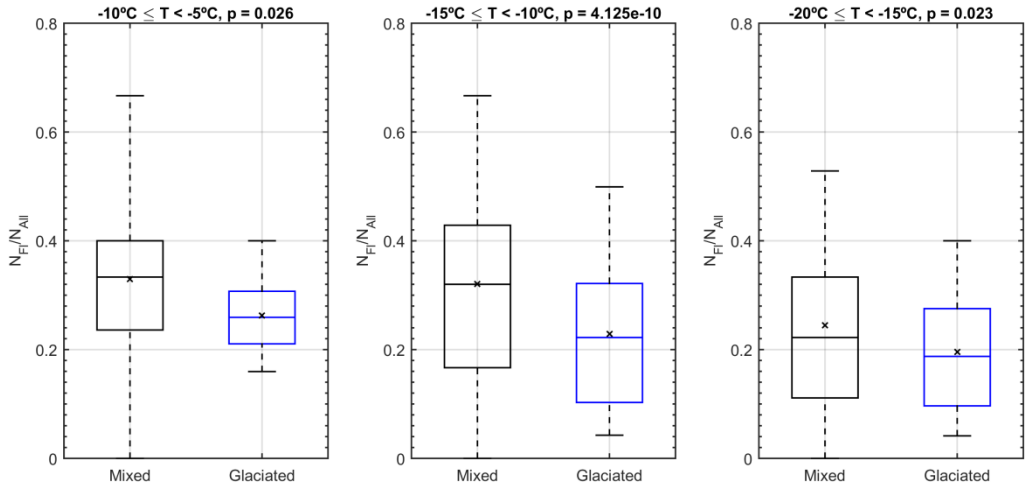

Figure 4. Influence of cloud type on fluorescent fraction for the studied temperature regimes.

Pg. 26074 Line 14: You bring up a point that may be worth discussing in detail further, in that certain cloud events had large fluctuations of fluorescent aerosol fractions while some do not.

> There is no obvious apparent reason for the large fluctuations of fluorescent aerosol fractions observed in some cloud cases. This may be an effect of sampling several different air masses during a single cloud event. We have now included this in the discussion.

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Interactive Comment by E. Herrmann

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

Dear authors / Hi guys!

With some interest I have followed the outcome of your latest Jungfraujoch campaign (this and the snow-blow paper). Referee #1 has already made some useful remarks (most of which I would agree with) but there is one more issue I'd like to raise. In figure 6, you show something that you call particle size distributions. These extend from ~250 nm to ~15 micrometer and have a peak between 400 and 800 nm or so. However, there is a bit of a problem with that: The actual size distributions at Jungfrauoch, measured behind the same inlet, do not look at all like the plots in figure 6 (see http://onlinelibrary.wiley.com/doi/10.1002/2015JD023660/full). The size distribution at Jungfraujoch has an accumulation mode at roughly 140 nm, beyond that is merely that mode's "tail" and the occasional dust particle from Africa.

The explanation for this odd result can be found earlier in the manuscript (page 26071 bottom) where you write that the WIBS doesn't provide reliable numbers for particles with diameters below 800 nm. With this in mind it would seem that figure 6 merely shows the WIBS signal. I could get behind that. But it is important to note that the WIBS signal is NOT the particle size distribution. Instead I would say that the WIBS signal is the actual size distribution multiplied with the WIBS detection efficiency curve. And that detection efficiency is significantly smaller than 1 below 800 nm (as the text implies) and appears to be 0 (zero) at 250 nm (as figure 6 suggests). It is therefore a bit misleading to write that figure 6 shows particle size distributions at Jungfraujoch. And I also want to stress that the actual size distribution does not have a mode at 580 nm (as stated in the manuscript). That "mode" is a result of WIBS detection efficiency. It would be nice if you would take this into account in the revision. Considering that not only the counting efficiency decreases but also the fluorescence measurement is unreliable below 800 nm, one of course has to wonder how meaningful the information in figure 7 is. You might want to discuss this in the revised manuscript as well

We thank the E. Herrmann for their helpful comments and recommendations. We agree that the sampling of particles with the WIBS is unreliable under 0.8 µm and we will revise the discussions of the size distributions to clarify these effects of the reduction in counting efficiency and include appropriate caveats.

We now include SMPS data in our revised analysis as part of a comparison to long term data sets which provides accurate size resolved concentrations down to D_p ~ 20 nm which we compare to the WIBS data to highlight the reduction in performance at small sizes. This can be found in our response to referee #1.

Beside this main point I wanted to make, some other things caught my eye. For example, what exactly is the motivation for discussing the SDE events separately? The current manuscript doesn't appear to be all that massive to make this necessary.

During the first SDE we sampled at a different location (Schilthorn) with the WIBS which makes directly linking the cloud microphysics measurements made at the jungfraujoch difficult as the two mountain sites are separated by several kilometres horizontally and approximately 500m vertically. Unfortunately during the second SDE we do not have reliable cloud microphysics data to perform analysis on due to artefacts resulting from the extremely high dust concentrations observed during the SDE.

And why do you call clouds glaciated that are still 50% water? While one certainly needs to define a threshold, 50% feels like an unintuitive choice.

> We have now revised the threshold between mixed phase and glaciated conditions to IMF ≥ 0.9 to be in keeping with Lloyd et al., (2015). A short discussion of the effect this has had on the analysis is provided in our response to referee #2.

Finally, the colours got mixed up in the legend of the top panel of figure 3 (the caption is correct).

> We thank E. Herrmann for bringing this to our attention and this will be corrected in the revised manuscript.

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Lloyd, G., Choularton, T. W., Bower, K. N., Gallagher, M. W., Connolly, P. J., Flynn, M., Farrington, R., Crosier, J., Schlenczek, O., Fugal, J., and Henneberger, J.: The origins of ice crystals measured in mixed-phase clouds at the high-alpine site Jungfraujoch, Atmos. Chem. Phys., 15, 12953-12969, doi:10.5194/acp-15-12953-2015, 2015.

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Date: 5 February 2016

Observations of fluorescent aerosol–cloud interactions in the free troposphere at the ~~Sphinx high Alpine research station,~~ High Altitude Research Station Jungfraujoch

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Abstract

The fluorescent nature of aerosol at a high altitude Alpine site was studied using a wide-band integrated bioaerosol (WIBS-4) single particle multi-channel ultra violet-light induced fluorescence (UV-LIF) spectrometer. This was supported by comprehensive cloud microphysics and meteorological measurements with the aims of cataloguing concentrations of bio-fluorescent aerosols at this high altitude site and also investigating possible influences of UV-fluorescent particle types on cloud–aerosol processes.

Analysis of background free tropospheric air masses, using a total aerosol inlet, showed there to be a minor ~~but statistically insignificant~~ increase in the fluorescent aerosol fraction during in-cloud cases compared to out of cloud cases. The size dependence of the fluorescent aerosol fraction showed the larger aerosol to be more likely to be fluorescent with 80 % of 10 µm particles being fluorescent. Whilst the fluorescent particles were in the minority (N_{Fl}/N_{All} = 0.27 ± 0.19), a new hierarchical agglomerative cluster analysis approach, (Crawford et al., 2015), revealed the majority of the fluorescent aerosol were likely to be representative of fluorescent mineral dust. A minor episodic contribution from a cluster likely to be representative of primary biological aerosol particles (PBAP) was also observed with a wintertime baseline concentration of $0.1 \pm 0.4\,L^{-1}$. Given the low concentration of this cluster and the typically low ice active fraction of studied PBAP (e.g. *pseudomonas syringae*) we suggest that the contribution to the observed ice crystal concentration at this location is not significant during the wintertime.

1 Introduction

The formation of cloud particles and their subsequent interactions with the atmosphere are highly uncertain, with the formation and evolution of mixed phase and glaciated clouds being poorly understood (Penner et al., 2001). Improving our understanding of primary ice nucleation is critical in underpinning these uncertainties as even modest concentrations of primary ice can result in the rapid glaciation via secondary ice production mechanisms

and subsequently cause precipitation in mixed phase clouds, drastically changing cloud lifetime (Lloyd et al., 2015; Crawford et al., 2012; Crosier et al., 2011). Many candidate aerosol , e.g. 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. Many candidate aerosols have been assessed for their heterogeneous ice nucleating ability with a particular emphasis being placed on mineral dust and primary biological aerosols. The ice nucleating efficiency of many naturally occurring and surrogate dust aerosols have been investigated and they are generally considered to be efficient ice nuclei with observations of ice activation occurring over water subsaturated and supersaturated conditions at temperatures below $-10\,^{\circ}C$ (Hoose and Möhler, 2012). The influence of accumulated coatings such as secondary organic aerosol, sulphuric acid and ammonium sulphate through atmospheric processing have also been assessed where it was found these act to significantly increase the saturation ratio required for ice nucleation, effectively deactivating an otherwise ice active mineral dust (Chernoff et al., 2010; Koehler et al., 2010; Sullivan et al., 2010; Cziczo et al., 2009; Eastwood et Saharan desert dust was observed during an experiment in a Florida region where it was suggested that the dust may have been acting as an effective high temperature ice nucleus nuclei resulting in the observed glaciation of an altocumulus cloud at $-5\,^{\circ}C$ (Sassen et al., 2003). Saharan desert dust was also found to be the major non-volatile component of ice crystal residuals in cirrus over the Alps (Heintzenberg et al., 1996). The high ice nucleation efficiency of mineral dusts and their capacity for long-range transport therefore make them a potentially potent significant component in the formation and modification of clouds worldwide. Certain Primary Biological Aerosol Particles primary biological aerosol particles (PBAP) exhibit the ability to nucleate ice and it has recently been suggested that ice active PBAP may have evolved over geological time scales to enhance rainfall, fostering an environment beneficial to the growth of plants and microorganisms through the so-called bioprecipitation feedback cycle (Morris et al., 2014). A small number of bacterial strains, fungal spores and rusts have been identified as ice active at temperatures warmer than $-10\,^{\circ}C$ due to the presence of an ice nucleating protein in the outer

cell wall which is structurally similar to ice, facilitating ice growth (Kajava and Lindow, 1993; Govindarajan and Lindow, 1988; Hoose and Möhler, 2012). However, of the ice active bacterial strains studied so far only a small fraction nucleates ice at very warm temperature, e.g. Möhler et al. (2008) demonstrated that *Pseudomonas syringae* have a maximum ice active fraction of 0.005 at $-9.7\,^{\circ}$C~~, however,~~ . However, they may still play a significant role in the formation and modification of ~~cloud as~~ clouds; plant surface derived bacterial aerosol can be transported to the higher levels of the atmosphere in high concentrations as a result of heavy rainfall and storm generated uplift (Crawford et al., 2014; DeLeon-Rodriguez et al., 2013; Huffman et al., 2013). The ~~Sphinx high Alpine research station~~ High Altitude Research Station Jungfraujoch has hosted several intensive measurement campaigns to study ~~cloud aerosol interactions~~ cloud-aerosol interactions (e.g., Targino et al., 2009; Choularton et al., 2008; Cozic et al., 2007; Verheggen et al., 2007). Previous measurements at the site have found there to be an enhancement of mineral dust in cloud particle residuals compared to interstitial aerosol measurements (Kamphus et al., 2010). This study also deployed a portable ice nucleation chamber during June 2009, where two Saharan Dust Events (SDE) were reported. During the SDE it was found that ice ~~nucleus~~ nuclei concentrations were correlated with larger aerosol ($~~D > 0.5~~ D_p > 0.5\,\mu$m) with reported deposition mode ice nuclei concentrations of up to several hundred per litre. This is discussed in more detail in the companion paper to this study by Lloyd et al. (2015). In this study, we present contemporaneous aerosol and cloud microphysics measurements at the same site to characterise the fluorescent constituents of aerosol and their possible role in cloud processes.

2 Methods

2.1 Site description

During January and February 2014, the Ice NUcleation Process Investigation And Quantification (INUPIAQ) project was conducted at the ~~high Alpine research station~~ High Altitude

Research Station Jungfraujoch (JFJ, 3580 m above sea level; 46.55° N, 7.98° E) in Switzerland to investigate the influence of a range of aerosol types on ice crystal number concentration alongside secondary ice processes in natural supercooled ~~cloud. The~~ clouds. The facility is situated on a mountain ridge in between the peaks of the Jungfrau and Mönch with the Great Aletsch Glacier, the largest in the Alps, to the south and is well away from major anthropogenic pollutions sources. The JFJ site is enveloped by cloud for approximately 37 % of the ~~year~~ time making it ideal for studying cloud–aerosol interactions, with the site residing in the free troposphere for most of the time during the wintertime ~~(Baltensperger et al., 1998)~~(Baltensperger et al., 1998; Herrmann et al., 2015).

2.2 Instrumentation and inlets

Fluorescent aerosol number size distributions were measured using a Wideband Integrated Bioaerosol Spectrometer (WIBS, Version 4, University of Hertfordshire) on a single particle basis and designed primarily for identifying biofluorophores. A full technical description can be found in Kaye et al. (2005), while various applications and analysis approaches including monitoring at high altitude sites can be found in ~~Gabey et al. (2013); Stanley et al. (2011) and Crawford et al. (2014)~~ Crawford et al. (2014) and Gabey et al. (2013); Stanley et al. (2011). A brief description of the instrument is now given. The WIBS-4 spectrometer exploits the principle of UV light induced fluorescence where a particle of interest is excited with UV radiation and the resultant fluorescence is detected, with fluorescence being an indicator that the particle may be biological. In the WIBS-4 aerosol is drawn into the sample volume and illuminated by a 635 nm laser and the resultant forward scattered light is used to determine the particle size and shape using a quadrant detector Kaye et al. (2005). Side scattered light is collected and sequentially triggers two xenon flash lamps, filtered to excite the sampled particle at 280 and 370 nm respectively. The first lamp is pulsed and the resultant fluorescence is collected, filtered and passed to two fluorescence detectors. The detectors are filtered to measure fluorescence over two detection bands (320–400 and 410–650 nm) which are then recorded. The second flash lamp is then triggered

and the fluorescence detected by the ~~2nd~~ second band is recorded. The whole process takes approximately 25 µs and the instrument has a maximum particle analysis rate of 125 particles s^{-1}. This provides three measurements of particle fluorescence over two excitation wavelengths, particle size and an approximation of particle shape, all on a single particle basis Kaye et al. (2005). The excitation and detection wavelengths have been selected to conform to ~~known~~ auto-fluorescence bands of common components of biological materials (e.g. proteins, tryptophan and Nicotinamide adenine dinucleotide, NADH, the latter related to cell metabolism) such that they can be discriminated from non-biological, non-fluorescent particles Kaye et al. (2005). Due to detector sensitivity and background fluorescence within the WIBS-4 optical chamber ~~both~~ the fluorescence of aerosol with diameters $D_p < 0.8\,\mu m$ cannot be accurately measured, and the counting efficiency decreases ~~, Gabey et al. (2011)~~(Gabey et al., 2011). Therefore the analysis presented here is limited to aerosols with diameters greater than 0.8 µm, unless otherwise stated. Whilst WIBS-4 instruments have many advantages over traditional UV-LIF spectrometers, limitations include difficulties in discriminating different classes of biological ~~classes~~ particles unambiguously and fluorescent non-biological aerosols must be identified. Fluorescence of some mineral dusts was examined by Pöhlker et al. (2012) who characterised their weak fluorescence properties allowing them to be generally discriminated from common PBAP using UV-LIF. In this study we use a new hierarchical agglomerative data processing method for WIBS-4 UV-LIF measurements to discriminate between particle types ~~.~~ and the methods used are described in section 4. A detailed discussion of this can be found in Crawford et al. (2015).

The WIBS-4 sampled from a total inlet (TI) which is now described. The TI samples all particles with $D_p < 40\,\mu m$ and for wind speeds $< 20\,m\,s^{-1}$. The sampled air is first heated to $+20\,^{\circ}C$, evaporating droplets and ice crystals such that their residuals are sampled along with any interstitial aerosol (Weingartner et al., 1999).

A custom built scanning mobility particle sizer (SMPS) has sampled continuously from the TI since 2008. It consists of a differential mobility analyzer (DMA, TSI 3071) and a condensation particle counter (CPC, TSI 3775) and it measures the aerosol

size distribution between 20 and 600 nm in diameter with 6 minute time resolution (Herrmann et al., 2015). This was used to determine the origin of the sampled air masses, using the concentration of paericles larger then 90 nm in diameter as desrcibed in section 3.1 and Herrmann et al. (2015).

Comprehensive cloud microphysics measurements were made at the site and are described in Lloyd et al. (2015). In this study~~measurements of~~ , cloud droplet and ice crystal number concentrations were measured respectively with a Cloud Droplet Probe (CDP-100, Droplet Measurement Technologies, DMT), described by Lance et al. (2010), and a 3 View – Cloud Particle Imager (3V-CPI~~, a multi-probe comprising a 2-D stereo imaging spectrometer (2D-S)and Cloud Particle Imager (CPI), e. g. Lawson et al. (2015).~~). The CDP-100 is an optical scattering spectrometer able to size particles in the range $2 < D_p < 50\,\mu m$ whilst the 3V-CPI is an integrated 2-D-Stereo (2DS) LED imaging spectrometer and Cloud Particle Imaging (CPI), CCD imaging spectrometer with resolutions of 10 and 2.3 µm respectively (Lawson et al., 2015). These are capable of measuring ice particle size distributions between 10–1280 µm and able to discriminate particle habit (based on shape analysis) for particles greater than approximately 25–30 µm. Details of the analysis techniques used for these instruments are provided in ~~Crosier et al. (2014); Lloyd et al. (2015)~~Crosier et al. (2014) and Lloyd et al. (2015).

3 Results

During the experiment, there were two extended ~~Saharan dust events~~ SDE's (00:00 CET, 1 February–00:00 CET, 2 February and 04:30 CET, 18 February–19:00 CET, 19 February). In this paper we focus on the period outside these events in order to characterise the behaviour of high Alpine fluorescent aerosol under typical winter time background conditions. Discussion of the SDE's will be described elsewhere. 5 min integration periods are used in all analysis unless otherwise stated.

3.1 Meteorological conditions

An overview of the meteorological conditions at the JFJ site over the background period 6–18 February is provided in Fig. 1. Average temperatures of -11.3 ± 4.3 and $-14.6 \pm 3.3\,^{\circ}\mathrm{C}$ were reported for out of cloud and in cloud periods respectively with wind speeds of $5.2 \pm 3.3\,\mathrm{m\,s^{-1}}$. Daily HYSPLIT back trajectory analysis (Fig. 2) showed the majority of air masses to have passed over the Atlantic ocean in the preceding 72 h during this period. Analysis of wind speed and direction shows the highest concentrations of fluorescent aerosols occur when the wind is coming from the south east for wind speeds in excess of $15\,\mathrm{m\,s^{-1}}$, i.e. coincident with flow up from the Aletsch glacier.

We use the approach of Herrmann et al. (2015) to determine the origin of the sampled air massess so that boundary layer influenced air massess can be excluded from analysis; here 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 $\mathrm{cm^{-3}}$ 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 $\mathrm{cm^{-3}}$. These values were found to be lower in winter so we use $N_{90} < 30$ $\mathrm{cm^{-3}}$ to be representative of background FT conditions and $N_{90} < 50$ $\mathrm{cm^{-3}}$ to be representative of "FT-like" conditions during the sampling period as described in Herrmann et al. (2015). A time series of the SMPS N_{90} concentration for the analysis period is presented in Fig. 3 where the background FT condition of $N_{90} < 30$ $\mathrm{cm^{-3}}$ is met 66.2% of the time and "FT-like" conditions where $N_{90} < 50$ $\mathrm{cm^{-3}}$ is met 88.4% of the time. Periods with $N_{90} > 50$ $\mathrm{cm^{-3}}$, such as the extended period between 09:00 15/02 - 09:00 16/0, are excluded from analysis.

3.2 Background observations of fluorescent aerosol

~~The period between the two SDE is considered~~

To assess the background conditions during the sampling period we have compared the aerosol data collected during the campaign to long term measurements made during February at the site between 2009 and 2014. Fig. 4 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 non-fluorescent and fluorescent size resolved concentrations where the SMPS reports mobility diameter and the OPC and WIBS report optical diameter. 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 background aerosol ~~concentrations at the site during the wintertime. Average~~ concentration at the Jungfraujoch during wintertime.

The average out of cloud total coarse aerosol, N_{All}, and total fluorescent aerosol concentrations, N_{Fl}, measured by the WIBS-4 were 30.6 ± 19.3 and $6.3 \pm 5.7\,\mathsf{L}^{-1}$ respectively for the period 6–18 February, as shown in Fig. 5 (5 min averages).

To investigate the potential interaction of fluorescent aerosol with clouds we have studied the fluorescent aerosol concentration fraction ($N_{\mathsf{Fl}}/N_{\mathsf{All}}$) over different temperature regimes for out of cloud, mixed phase and glaciated conditions as summarised in Fig. 6. Here we define out of cloud as all periods where the total water content (TWC) is less than $0.01\,\mathsf{g\,m}^{-3}$; mixed phase as all periods where the TWC $\geq 0.01\,\mathsf{g\,m}^{-3}$ and ice mass fraction (IMF) is less than ~~0.5~~0.9; and glaciated as all periods where TWC $\geq 0.01\,\mathsf{g\,m}^{-3}$ and IMF ~~≥ 0.5. It~~ ≥ 0.9.

To test the statisitical significance of these results we have performed a 1 way ANOVA analysis on subsets of the data which we now describe; first we assessed the influence of temperature separately for in cloud (TWC $\geq 0.01\,\mathsf{g\,m}^{-3}$) and out of cloud (TWC $< 0.01\,\mathsf{g\,m}^{-3}$) conditions where it can be seen in Fig. 6 that in each case the fluorescent fraction decreases with decreasing temperature. The ANOVA analysis returns small p-values (4×10^{-6} and 1×10^{-4} for the out of cloud and in cloud cases respectively) which indicates that the means are statistically significantly different; however, the spread in values are large; next we assessed the influence of the presence of cloud on fluorescent

fraction at each temperature by comparing the out of cloud and in cloud cases for each temperature regime. This shows that the fluorescent fraction is generally increased in clouds (Fig. 7, top panels) with p-values indicating that the means are significantly different ($p < 0.05$); finally we assessed the influence of cloud type on the fluorescent fraction for each temperature regime as shown in Fig. 7. Here it can be seen that the fluorescent fractions are generally greater in mixed phase conditions than in glaciated conditions.

In summary it can be seen across all temperature regimes that the average in-cloud fluorescent aerosol fractions were slightly greater ~~(~ 0.28, all temperatures)~~ than for out of cloud conditions ~~(~ 0.24, all temperatures), however the variations are large and the mean values of each case lie within one standard deviation of each other with no one case proving to be statistically significant. It was also observed that the~~ with the largest increase occuring during mixed phase conditions. The observed increase in the fluorescent aerosol fraction in mixed phase conditions is generally a result of a reduction in the non-fluorescent aerosol concentration relative to the corresponding out of cloud cases, rather than an enhancement in the fluorescent aerosol concentration. One possible explanation for this is that non-fluorescent aerosol has been removed via CCN activation and lost in precipitating raindrops in mixed phase clouds as this is not pronounced in the glaciated cases, however, caution must be applied when interpreting the results of this general approach as the differences in fluorescent aerosol fraction ~~decreases with decreasing temperature.~~ may be caused by differences in the sampled air masses for each case.

~~Figure~~Fig. 8 shows the fluorescent aerosol fraction for cloud events persisting for a minimum of 30 min in duration with mean, minimum and maximum observed average fluorescent aerosol fractions of 0.27 ± 0.12, 0.05 and 0.49 respectively over ~~34~~ 31 separate cloud events. It can be seen that many of the clouds feature large variations in fluorescent aerosol fraction, while others have relatively little variation ~~.~~ which may be an effect of sampling several different air massess during a single cloud event. The correlation between mean and median fluorescent aerosol fraction and the following meteorological and cloud microphysical parameters were investgated; 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 Fig. 9, along with the corresponding r^2 value where no significant correlation between parameters is observed. No apparent trend is observed between the ~~mean~~ fluorescent aerosol fractions and contemporaneous mean meteorological or cloud microphysical parameters, suggesting that particle fluorescence does not impact cloud evolution or formation.

The majority of cloud events occur in the $-15 \leq T < -10\,^{\circ}\mathrm{C}$ regime: Fig. 10 shows the average fluorescent and non-fluorescent particle size distributions for out of cloud, mixed phase and glaciated conditions in this temperature regime. In each case the single mode of the distribution occurs at 0.58 µm. ~~Figure~~, however, the counting efficiency for particles $D_P < 0.8\,\mu\mathrm{m}$ is low (Gabey et al., 2011) so the true mode is likely to be much smaller when measured with e.g. an SMPS as indicated in Fig. 4.

Fig. 11 shows the size dependence of the fluorescent aerosol fraction for the three studied temperature regimes for out of cloud, mixed phase and glaciated conditions. In each case, it was observed that the fluorescent aerosol fraction increases with size, with approximately 80 % of 10 µm particles being fluorescent in nature, with the fluorescent aerosol fraction decreasing to approximately 20 % for 1 µm particles. Caution must be applied when interpreting the sub-micron fluorescent aerosol fraction due to the reduced fluorescent counting efficiency for particles $D_P < 0.8\,\mu\mathrm{m}$ (Gabey et al., 2011) which may lead to an underestimation of the fluorescent aerosol fraction at small sizes. For clarity and ease of comparison only the mean ratios for each case are presented here. Individual plots for each case showing the mean and standard deviation of the fluorescent ratio are provided as supplementary material.

4 Analysis of fluorescent aerosol characteristics

To probe the nature of the fluorescent aerosols the single particle data from the period 6–18 February (approximately 27 000 fluorescent particles) was clustered using the Ward hierarchical agglomerative cluster analysis linkage and Z score normalisation technique

with the log of the ~~size~~ diameter and particle asymmetry factors (AF) used to improve the symmetry of the cluster distribution. For further details on the hierarchical agglomerative cluster analysis method used here see Crawford et al. (2015). The Calinski–Harabasz metric was used to determine the optimum cluster solution to retain, returning a 3 cluster solution as shown in Fig. 12. Clusters 1 and 2 were the dominant clusters, both of which display weak fluorescence which is characteristic of mineral dust (Pöhlker et al., 2012). The sum of particle concentrations from both clusters 1 and 2 correlated well with the total fluorescent particle concentration ($(N_{cl1+cl2}) = 0.3 + 0.94 \times N_{Fl}$, $r^2 = 0.99$) with campaign average concentrations of 1.7 ± 3.3 and 4.9 ± 8.8 L^{-1} respectively. Cluster 3 displayed significantly higher fluorescence in all 3 channels suggesting that this was likely representative of biological material (Crawford et al., 2014). However, periods during which cluster 3 particles appeared were sparse with typical average concentrations over the period of $0.1 \pm 0.4\,L^{-1}$ observed. Very occasional episodic events with maximum concentrations reaching ~~of~~ the order of a few per litre were observed. We would expect ~~lower~~ low concentrations of local PBAP in the wintertime at this site due to reduced surface sources of seasonal PBAP coupled with an annual minimum in planetary boundary layer (PBL) height ~~which would allow for very little in the way of PBL influenced air to reach the research station (Ketterer et al., 2014; Collaud Coen et al., 2011; Nyeki et al., 1998). In summary~~ (Ketterer et al., 2014; Collaud Coen et al., 2011; Nyeki et al., 1998).

In summary, the majority of fluorescent aerosol sampled at the site during these periods is likely non-biological in nature with only minor episodic contributions from bioaerosols. Such low concentrations of PBAP are unlikely to have any significant impact on cloud evolution through ~~ice nucleation~~ primary ice nucleation alone due to the low ice active fractions reported for typical PBAP; e.g. ~~assuming the cluster is~~ if the cluster was representative of *Pseudomonas syringae* (Möhler et al., 2008) this would yield an IN concentration of only $5 \times 10^{-4}\,L^{-1}$ which is several orders of magnitude less than the reported ice crystal concentration (Lloyd et al., 2015)~~.~~, however, we can only speculate on the source of this cluster and this is used as an illustrative example only. Low concentrations of primary ice may cause glaciation via secondary mechanisms such as the Hallet-Mossop (HM)

process and Wegener-Bergeron-Findeisen (WBF) process (e.g. Crawford et al. (2012)) which we now discuss in relation to this study; In this study secondary ice production via the HM process was ruled out as the clouds observed were rarely within the active temperature range for this process as discussed in the Lloyd et al. (2015) companion study; a second companion study by Farrington et al. (2015) investigated the potential influence of the WBF process at the site where they found that the critical updraft speed (as defined by Korolev and Mazin (2003) and Korolev (2007)) to maintain mixed phase conditions was less than the observed updraft velocity for the majority of the INUPIAQ campaign using the ice 2D-S size distribution as the input for the $N_i r_i$ term, as such they concluded that glaciation via the WBF process was not significant. Reducing the $N_i r_i$ term to the typical bio-IN primary ice concentrations observed would reduce this critical threshold such that it would be significantly less than the observed updrafts.

5 Summary and conclusions

Analysis of 288 h of contemporaneous aerosol fluorescence and cloud microphysics measurements made during wintertime background conditions at a high Alpine site revealed that the majority of aerosol sampled with a WIBS-4 UV-LIF spectrometer were non-fluorescent with only 27 % of the aerosol displaying fluorescence. We investigated the potential links between aerosol fluorescence and cloud microphysics both in general and for ~~34~~ 31 individual cloud events persisting for at least 30 min and we report that there was no apparent link between the fluorescent aerosol fraction and observed cloud microphysical parameters and meteorology, suggesting that aerosol fluorescence ~~does~~ did not influence cloud formation/evolution at the site during the measurement period.

We observed that particle fluorescence is a strong function of size with 80 % of 10 µm particles displaying fluorescence, decreasing to 20 % at 1 µm. Hierarchical agglomerative cluster analysis of the fluorescent particles yielded a three cluster solution: two of the clusters displayed fluorescent characteristics consistent with fluorescent mineral dust and these clusters accounted for approximately 95 % of the observed fluorescent particles. The re-

maining cluster was moderately fluorescent in all three channels and is assumed to be biological in origin. Concentrations of the assumed PBAP cluster were sparse, occurring in occasional minor episodes with a baseline concentration of $0.1 \pm 0.4\,\mathrm{L}^{-1}$. Given the low concentration of this cluster and the typically low ice active fraction of studied PBAP (e.g. *Pseudomonas syringae*, Möhler et al., 2008) we suggest that the contribution to the observed ice crystal concentration at this location is not significant during the wintertime~~, however, analysis~~ . Analysis of wind speed and direction suggests that large emissions from sources from the Po valley region may advect up the Aletsch glacier during periods of high wind speed which may be of significance during the summer when the PBL is higher. We suggest that longer term data sets are required to examine this in detail.

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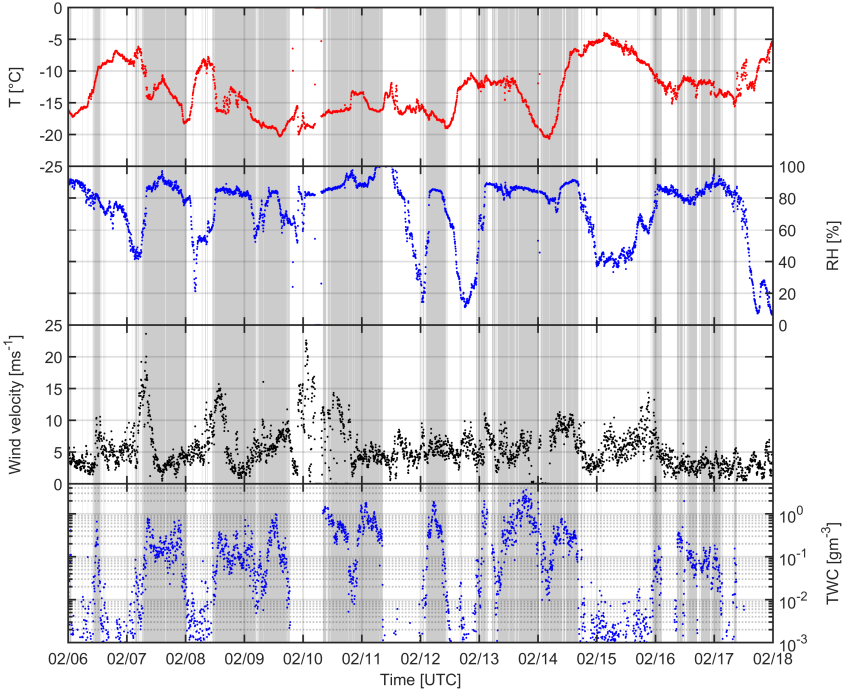

Figure 1. Time series of meteorological data and total water content at the JFJ site for the period 6–18 February. Grey shaded areas denote in cloud periods (TWC ≥ 0.01 g m^{-3}).

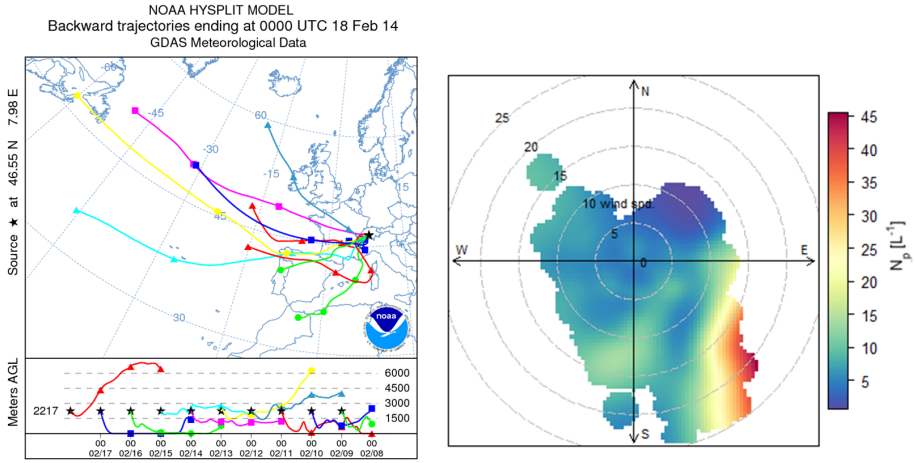

Figure 2. Left panel: HYSPLIT back trajectories for the period 8–18 February. Right panel: fluorescent aerosol concentration (L^{-1}) dependence on wind speed and direction. Wind speed denoted by concentric rings (5 m s^{-1} per ring).

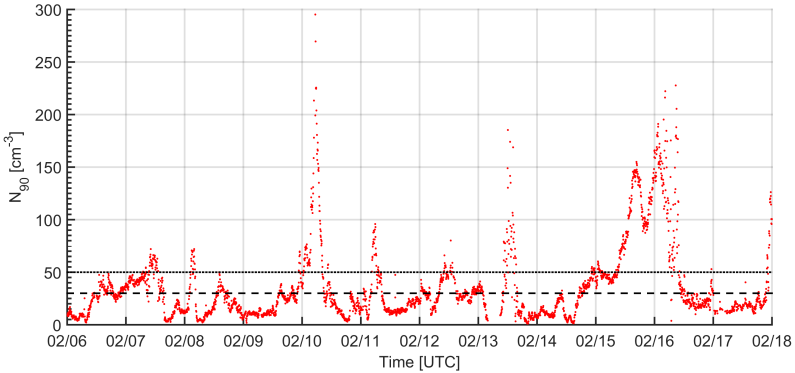

Figure 3. ~~Top panel: time~~ Time series of ~~total fluorescent, N_{FI}, (red) and total non-fluorescent, N_{NonFI}, (black) aerosol concentrations measured with~~ N_{90} concentration for the ~~WIBS-4 sampling from~~ analysis period. Dashed line denotes the ~~total inlet (TI)~~30 cm^{-3} background concentration described in Herrmann et al. ~~Middle panel: liquid (cyan~~2015)~~and ice (blue) water contents measured with~~ ; the ~~CDP-100 and 3V-CPI-2DS~~dotted line denotes this 50 cm^{-3} threshold used to distinguish free tropospheric conditions.~~Bottom panel: temperature.~~

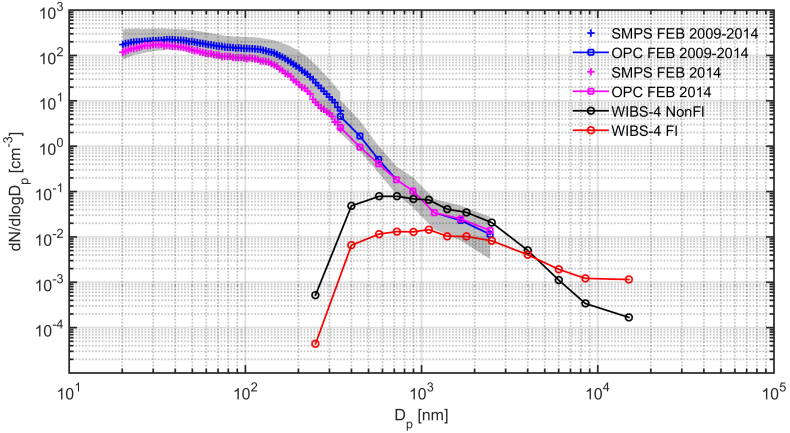

Figure 4. Comparison of long term median SMPS and OPC number size distribution measurements made during February 2009 to 2014 to those made during the 2014 campaign. Grey shaded area represents the quartiles of the long term measurements.

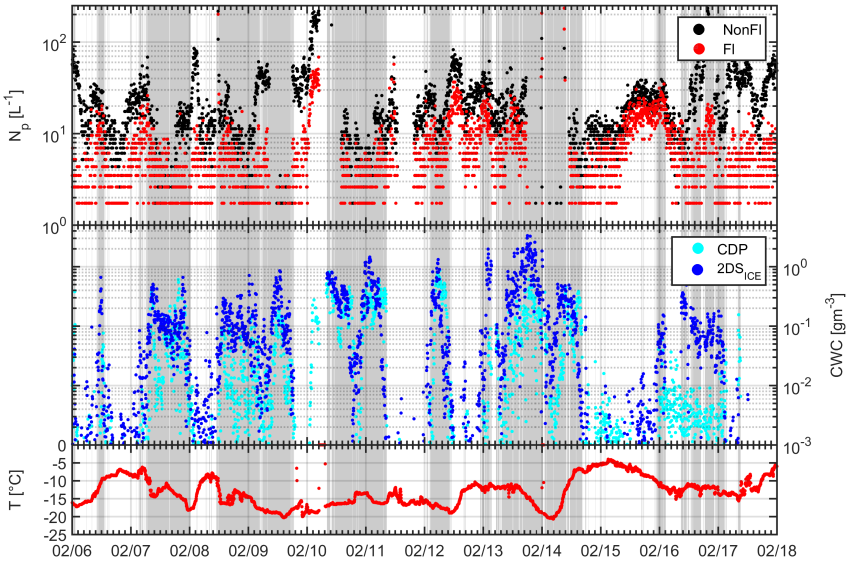

Figure 5. Top panel: time series of total fluorescent, N_{FI}, (red) and total non-fluorescent, N_{NonFI}, (black) aerosol concentrations measured with the WIBS-4 sampling from the total inlet (TI). Middle panel: liquid (cyan) and ice (blue) water contents measured with the CDP-100 and 3V-CPI-2DS. Bottom panel: temperature. Box and whiskers denote 5th, 25th, 50th, 75th and 95th percentiles. Grey shaded areas denote in cloud periods (TWC $\geq 0.01\,\mathrm{g\,m^{-3}}$).

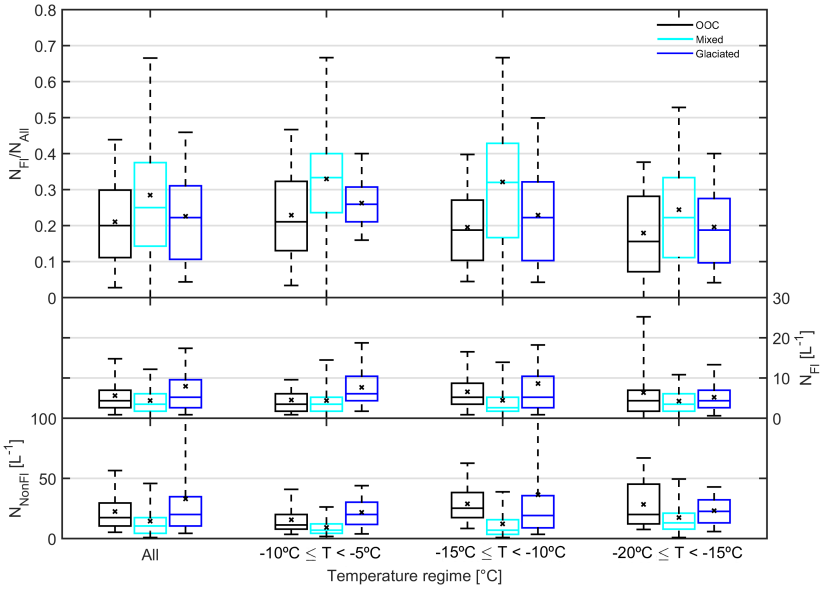

Figure 6. ~~Fluorescent~~ Top panel: fluorescent to total aerosol concentration ratio for out of cloud (black, TWC $< 0.01\,\mathrm{g\,m^{-3}}$), mixed phase (cyan, TWC $\geq 0.01\,\mathrm{g\,m^{-3}}$ and IMF ~~< 0.5~~ < 0.9) and glaciated (blue, TWC $\geq 0.01\,\mathrm{g\,m^{-3}}$ and IMF ~~≥ 0.5~~ ≥ 0.9) conditions sampled with the total inlet. Middle and bottom panels: total fluorescent, N_{FI}, and total non-fluorescent, N_{NonFI}, aerosol concentrations. Box and whiskers denote 5th, 25th, 50th, 75th, and 95th percentiles. x marker denotes mean.

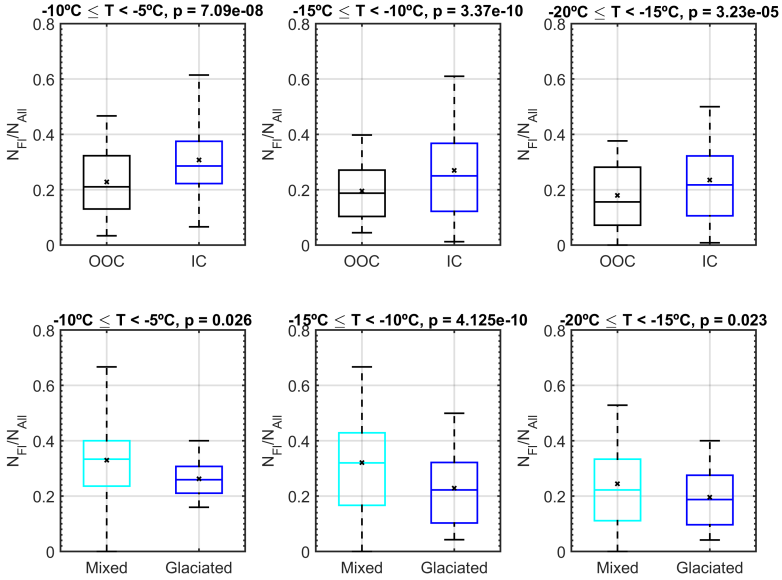

Figure 7. ~~Box and whisker plots~~ Top panels: Influence of cloud on fluorescent ~~to total aerosol concentration ratio~~ fraction for the studied temperature regimes. Bottom panels: Influence of cloud ~~events persisting~~ type on fluorescent fraction for ~~at least 30 in duration with accompanying~~ the studied temperature regimes. Box and whiskers denote 5th, ~~total water content~~ 25th, 50th, 75th, and ~~ice mass fraction measurements~~95th percentiles. x marker denotes mean. AVOVA 1 way p-values indicated at top of each panel.

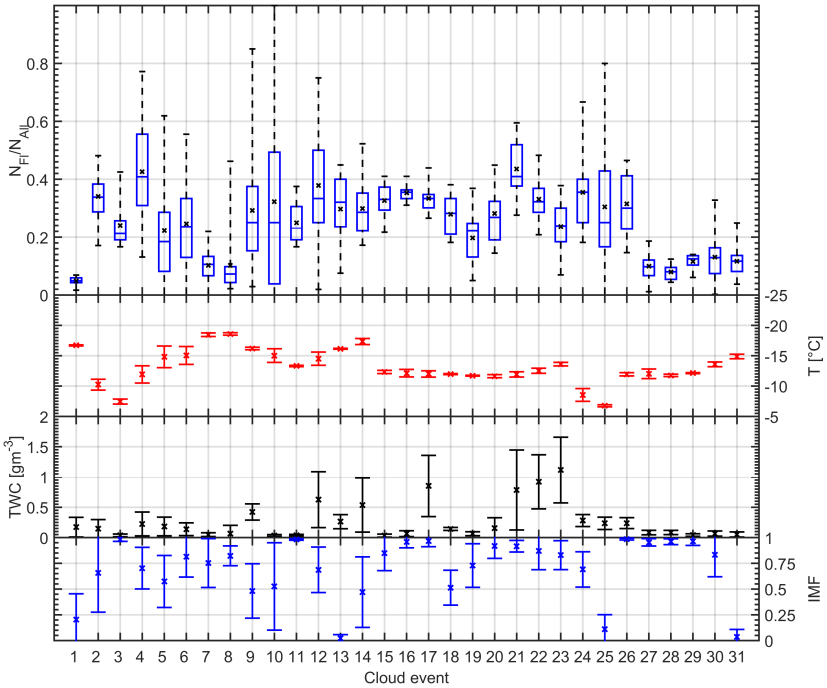

Figure 8. Box and whisker plots of fluorescent to total aerosol concentration ratio for cloud events persisting for at least 30 min in duration with accompanying temperature, total water content and ice mass fraction measurements. Box and whiskers denote 5th, 25th, 50th, 75th, and 95th percentiles. x marker denotes mean.

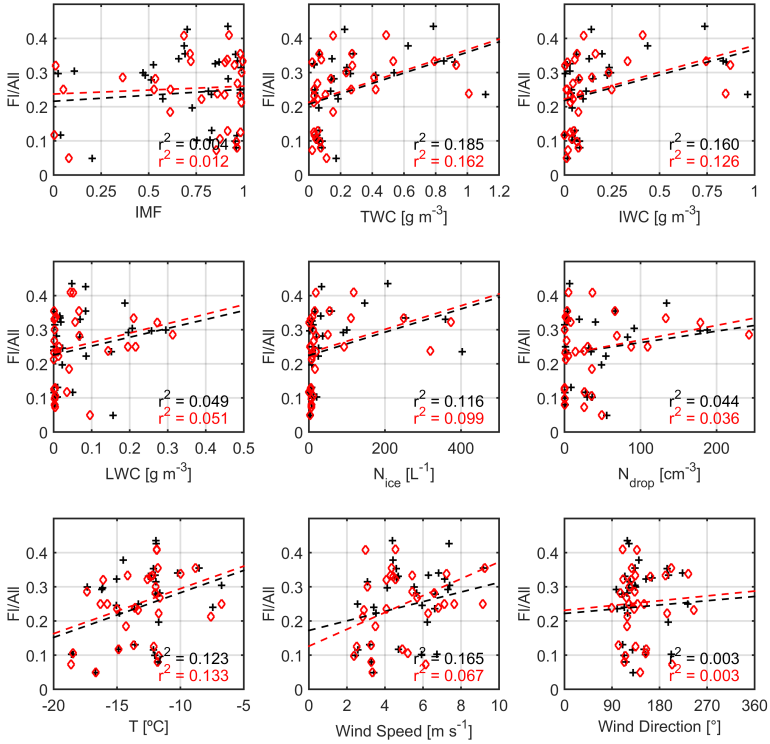
FI/All
IMF
r2 = 0.004
r2 = 0.012
TWC [g m-3]
r2 = 0.185
r2 = 0.162
IWC [g m-3]
r2 = 0.160
r2 = 0.126
LWC [g m-3]
r2 = 0.049
r2 = 0.051
Nice [L-1]
r2 = 0.116
r2 = 0.099
Ndrop [cm-3]
r2 = 0.044
r2 = 0.036
T [ºC]
r2 = 0.123
r2 = 0.133
Wind Speed [m s-1]
r2 = 0.165
r2 = 0.067
Wind Direction [°]
r2 = 0.003
r2 = 0.003

Figure 9. ~~Non-fluorescent~~ Correlation scatter plot of the fluorescent aerosol fraction to ice mass fraction (~~top~~IMF)~~and fluorescent particle size distributions~~ ; total water content (~~bottom~~TWC)~~for~~ ; ice water content (~~left to right~~IWC)~~out of cloud~~ ; liquid water content (~~OOC~~LWC)~~, mixed phase~~ ; ice crystal and ~~glaciated conditions over the~~ droplet number concentrations; temperature~~range $-15 < T \leq -10$~~ ; wind speed and direction for cloud events persisting for at least 30 min in duration. ~~Solid line is mean, dashed line is~~ Mean values are denoted by black + symbols and median values by red diamonds.~~5th–95th percentiles and interquartile range shown with light and dark grey areas respectively.~~

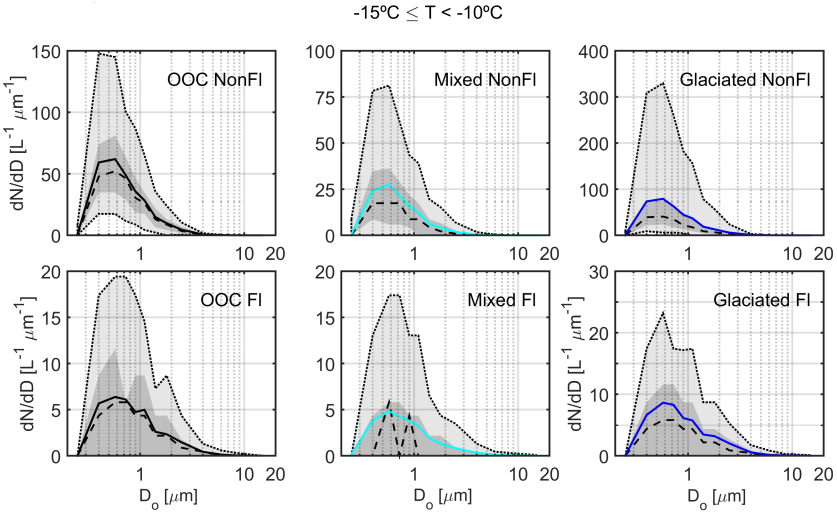

Figure 10. WIBS Non-fluorescent (top) and fluorescent particle size distributions (bottom) for (left to right) out of cloud (OOC), mixed phase and glaciated conditions over the temperature range -15 °C ≤ T < -10 °C. Solid line is mean, dashed line is median. 5th-95th percentiles and interquartile range shown with light and dark grey areas respectively.

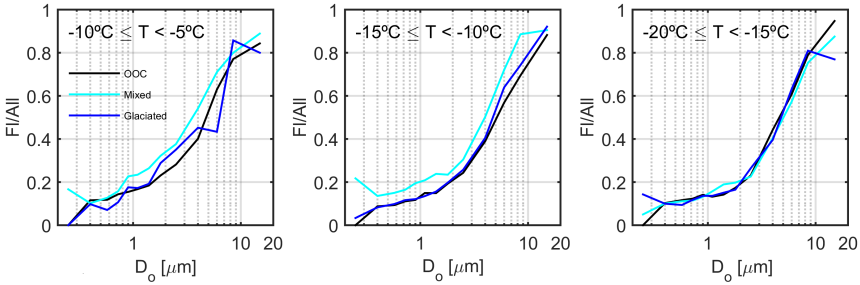

Figure 11. Size dependent fluorescent aerosol fractions for (left to right) out of cloud (black), mixed phase (cyan) and glaciated conditions (blue) over the three different temperature regimes studied.

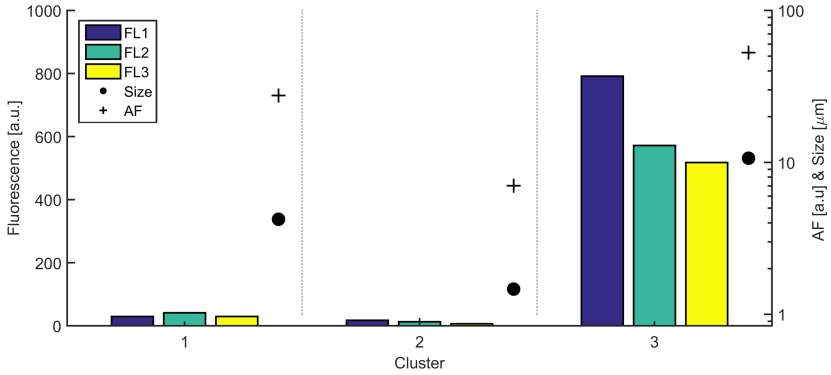

Figure 12. Mean cluster centres for the 3 cluster solution using Ward linkage and Calinski–Harabasz metric. Clusters contribute 25, 70 and 5 % respectively to the fluorescent particle population.