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:

Korolev, A. (2007). Limitations of the Wegener-Bergeron-Findeisen mechanism in the evolution of mixed-phase clouds. Journal of the Atmospheric Sciences, 64(9), 3372-3375.

Phillips, V. T. J., Choularton, T. W., Illingworth, A. J., Hogan, R. J., Field, P. R. (2003). Simulations of the glaciation of a frontal mixedâA˘ Rphase cloud with the Explicit Mi-˘crophysics Model. Quarterly Journal of the Royal Meteorological Society, 129(590),1351-1371.

Diehl, K., Wurzler, S. (2010). Air parcel model simulations of a convective cloud: Bacteria acting as immersion ice nuclei. Atmospheric Environment, 44(36), 4622-4628. Zeng, X., Tao, W. K., Zhang, M., Hou, A. Y., Xie, S., Lang, S., … Simpson, J. (2009). An indirect effect of ice nuclei on atmospheric radiation. Journal of the Atmospheric Sciences, 66(1), 41-61.

Pratt, K. A., DeMott, P. J., French, J. R., Wang, Z., Westphal, D. L., Heymsfield, A. J., … Prather, K. A. (2009). In situ detection of biological particles in cloud ice-crystals. Nature Geoscience, 2(6), 398-401.

Cantrell, W., Heymsfield, A. (2005). Production of ice in tropospheric clouds: A review. Bulletin of the American Meteorological Society, 86(6), 795-807.

Mossop, S. C. (1985). The origin and concentration of ice crystals in clouds. Bulletin of the American Meteorological Society, 66(3), 264-273.

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.

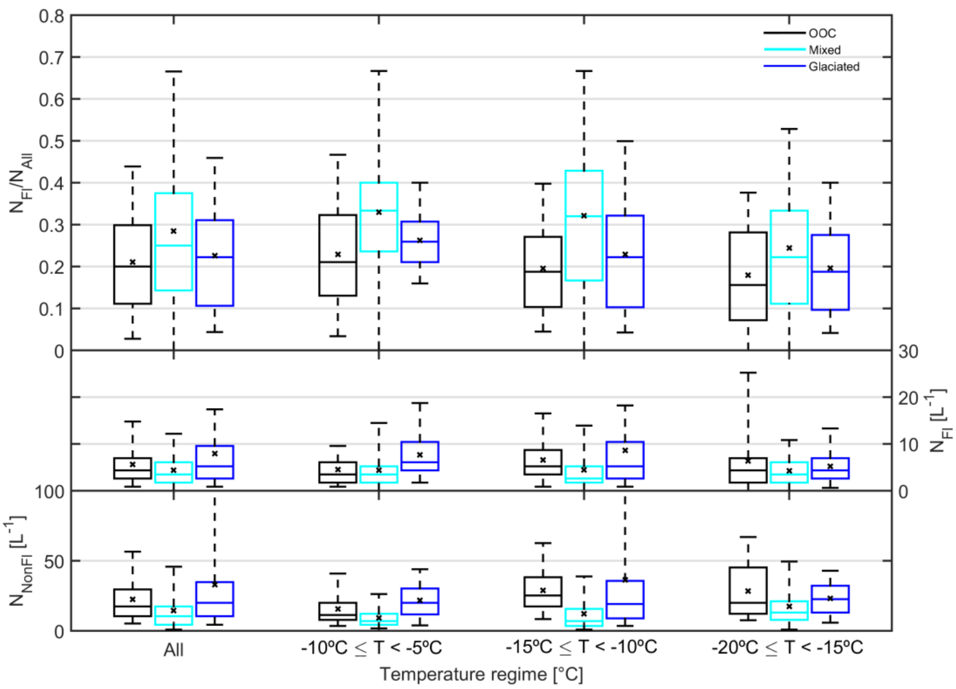

Figure 1. Revised figure 4. Now includes box and whisker plots showing the fluorescent and non-fluorescent aerosol concentrations for each case

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.

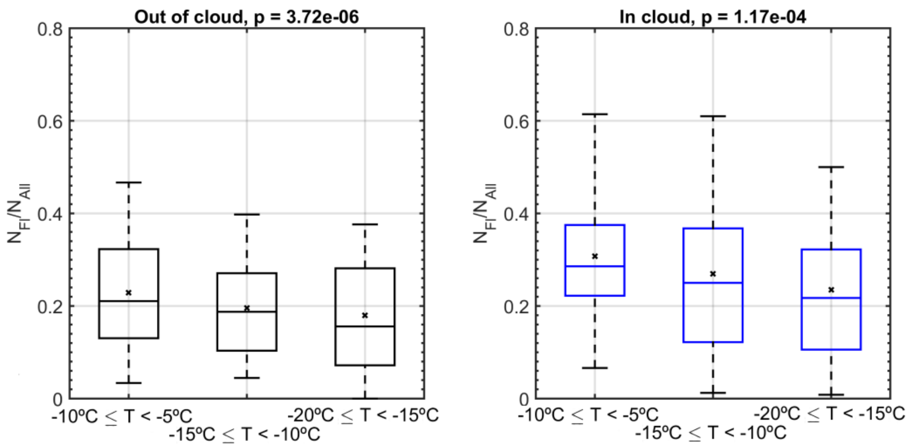

Figure 2. Influence of temperature on fluorescent fraction for out of cloud and in cloud cases.

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.

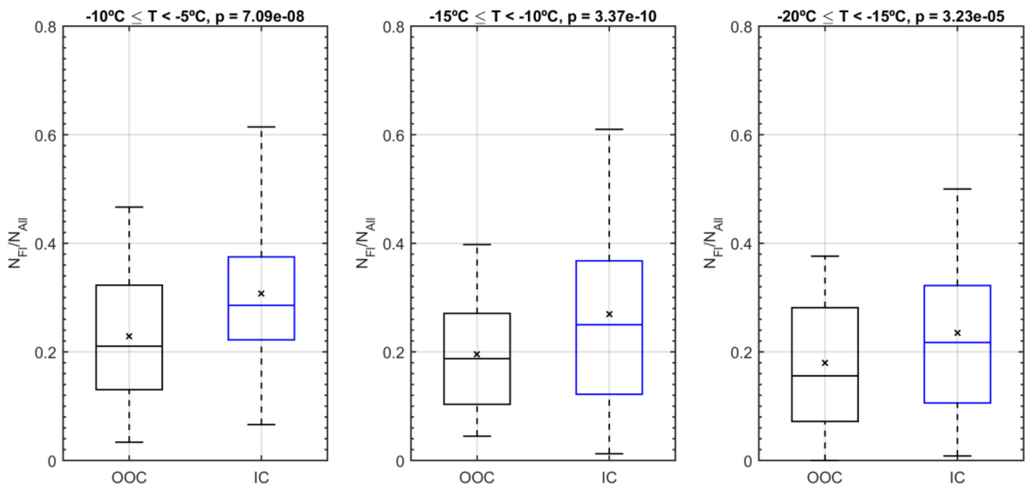

Figure 3. Influence of cloud on fluorescent fraction for the studied temperature regimes.

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.

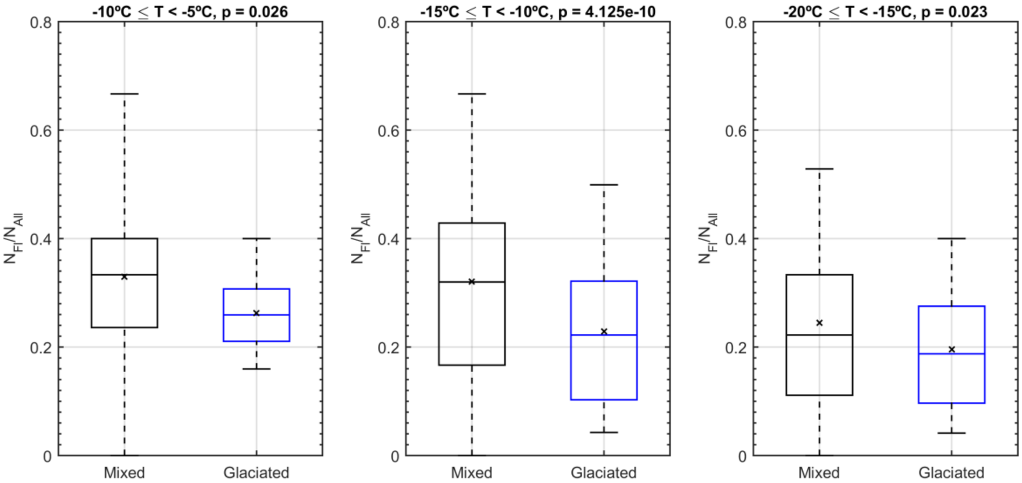

Figure 4. Influence of cloud type on fluorescent fraction for the studied temperature regimes.

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.

References

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Herrmann, E., et al. (2015), Analysis of long-term aerosol size distribution data from Jungfraujoch with emphasis on free tropospheric conditions, cloud influence, and air mass transport, J. Geophys. Res. Atmos., 120, 9459–9480, doi:10.1002/2015JD023660

Korolev, A. V. and Mazin, I. P.: Supersaturation of Water Vapor in Clouds, J. Atmos. Sci., 60, 2957–2974, doi:10.1175/1520-0469(2003)060<2957:SOWVIC>2.0.CO;2, 2003.

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Möhler, O., Georgakopoulos, D. G., Morris, C. E., Benz, S., Ebert, V., Hunsmann, S., Saathoff, H., Schnaiter, M., and Wagner, R.: Heterogeneous ice nucleation activity of bacteria: new laboratory experiments at simulated cloud conditions, Biogeosciences, 5, 1425–1435, doi:10.5194/bg-5-1425-2008, 2008. 26069, 26075, 26076