

### Anonymous Referee #3

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

This manuscript presents measurements of fluorescent aerosol made in Antarctica over a period of weeks using a WIBS. Although the authors find that fluorescent particles are a minor component (a few %) of total aerosol, there are some interesting features in the data worthy of publication. WIBS data is analyzed using a clustering method previously published by this group and 4 component populations are identified. Two of these clusters (together accounting for >97% of the fluorescent aerosol) are only weakly fluorescent and are hypothesized to be non-biological fluorescent aerosol, possibly dust. The other two clusters have more fluorescent intensity, are hypothesized to be biological and one of these is very similar to a cluster identified from laboratory samples of various pollen. Fluorescent loadings are analyzed as a function of wind speed for specific periods of interest and the authors state that high levels of fluorescent aerosols were primarily (though not always) associated with flow from the NE. Back trajectories are also analyzed and the authors posit that fluorescent aerosol (and thus pollen) arrives at the site as a result of long range transport from as far away as South America.

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

#### Comments:

This paper presents the first fluorescent aerosol observations reported for Antarctica and, as such, it is a worthy contribution to the literature. However I found portions confusing and also recommend including more information in certain places. Much of my discomfort arises from the fact that the 5 periods of interest seem rather arbitrary, at least given the information presented. As such, I don't know how to interpret observed differences between these periods or what they mean for fluorescent aerosol in Antarctica more generally. Specific suggestions for improvement are included below.

1. With any discussion of intensity of fluorescent signals the question of calibration arises. I appreciate that there is, as yet, no widely-accepted calibration for fluorescence in the WIBS and it seems that this instrument has been used in numerous laboratory and field studies without significant intentional modification. There is some discussion of this in the discussion of pollen identification (i.e. that the same instrument was used to look at pollen samples and they look very similar in intensity to Cl4) however it would be appropriate to include a more thorough related discussion in the methods section. Do the authors have any information regarding the stability of fluorescent intensity measurements over time? Are the instrument gains used here the same as those used in previously published work from this group? Can you comment on whether or how changes or instability in fluorescent sensitivity would affect the clustering algorithm? Can the authors comment on what kinds of laboratory-generated particles they have observed to fall into the weakly, moderately, medium and highly fluorescent populations? Those categories seem arbitrary and are used only minimally in the subsequent analysis.

Details of the sampling methodology are provided in Ruske et al. (2016). We will include a short description of the methodology in Appendix A where this is discussed.

The instrument used here is periodically sent back to the manufacturer for servicing, where the PMT voltages and xenon powers are noted. No significant changes in these values have been recorded between servicing. In the field prior to the start of measurement, instrument response was checked with fluorescent doped PSLs to verify the instrument is responding sensibly, however, absolute comparison between calibrations is not possible due to variation in fluorescent intensity between batches and the degradation of the doping material with time.

The version of the WIBS used here does not feature multiple gain modes and the detector gain is expected to be similar to that of previous studies.

It is not anticipated that the unsupervised clustering algorithm used here would be sensitive to differences in detector gain/fluorescent instability since absolute values are not referenced to a training dataset as would be the case with supervised methods.

The use of weakly/moderately/highly fluorescent is used as a descriptor to aid the discussion of classification. Generally from our laboratory characterisation we observe pollens to be highly fluorescent; fungal spores to be medium to highly fluorescent; bacteria to be moderate to medium fluorescent and mineral dusts to be weakly fluorescent. Savage et al. (2017) have recently performed a series of systematic laboratory characterisations which demonstrate that these particle types display a comparable broad trend in fluorescent spectra intensities.

2. On a related note, it would be good to include the numbers of particles sampled that fell in each cluster and also the number of particles that saturated the detector. Do the detectors for this WIBS saturate around 2000 counts? If so, given that the stated average intensity in the pollen population is  $\sim 1800 \pm 300$  after exclusion of saturating particles, it seems that a substantial fraction of pollen particles would saturate and you might be underestimating the contribution of that population.

We will include the number of particles attributed to each cluster in the revised manuscript.

There is an error in the technical description of the data analysis methods. In this analysis we retain any saturating particles to maximise the PBAP populations. We will correct this error in the revised manuscript and add a short discussion about why we have chosen to retain the saturating particles to maximise PBAP count.

3. A relatively minor point but, in your discussion of asymmetry factor, I believe dust is typically quite fractal (e.g. Bi, Huang et al, ACP 2016 or Yu, Zhu, et al, ACP 2015) yet your dust cluster AF indicates relatively sphere-like. Can the authors provide information to bolster confidence in the retrieved AF from the WIBS? (i.e. any data from calibrations with known aspherical particles or any corroborating reports of relatively spherical dust?)

The simple quadrant detector used here is incapable of detecting such fine detail, which may be captured by a more sophisticated detector such as the dual CMOS array used in the MBS for example. From our own laboratory characterisation experiments we have found that mineral dusts exhibit asymmetry factors of around 10, however, this work has not been published. Savage et al., (2017) performed a systematic characterisation of many particle types of interest using a WIBS-4A, which features a similar quadrant detector to the instrument used in this study. They characterised 13 mineral dusts of which many had an AF of approximately 10.

The quadrant detector AF proxy was calibrated for rod like particles using elliptical haematite as described in Kaye et al., (2007)

4. I am confused by the discussion surrounding the wind events. First, the authors define a level above which they consider fluorescent concentrations elevated and imply that they are going to look at periods where that happened. Then, however, two of the five periods in table 2 don't have elevated fluorescent concentrations (the 2nd and the 4th) while there are periods that seem to have elevated fluorescent concentrations that are not included in the analysis (i.e. early on in the project and on 11/29). Is the selection driven mainly by wind speed and direction? Why include the 5th period and not periods from 20-21 and 29 Nov? Are these just meant to be case studies of the different combinations of wind and aerosol loadings observed? Please clarify how these 5 periods of interest were chosen. It would also be helpful if these periods were marked in Figure 2 so that the reader doesn't have to mentally combine the table and the figure.

First we chose wind event A, based on its high concentrations of fluorescent material and PBAP cluster as a period of significant interest. This period featured high wind speeds from the NE, which is characteristic of the site (e.g., Renfrew & Anderson, 2002; Van Lipzig, et al., 2004) and confirmed by our own meteorological measurements during the experiment, as shown below:

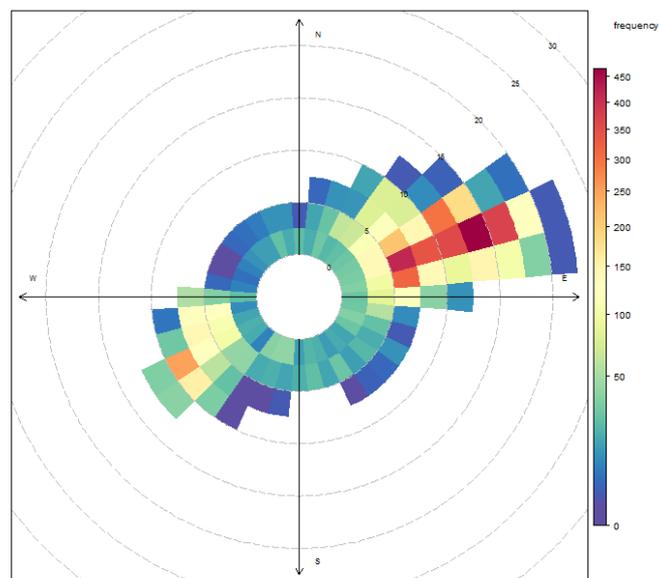


Figure 1. Polar histogram of wind speed and direction during MAC measurement period. Frequency indicates the number of 5 minute integrations. Rings indicate  $5\text{ms}^{-1}$  wind speed intervals.

Wind events B, C & D were chosen for comparison to event A as they have similar speeds and directions, yet the fluorescent and PBAP concentrations were significantly less than for event A. Event E was chosen as a case study demonstrating the much less frequent SW. We will clarify the selection criteria in the revised manuscript and include a shaded area highlighting the events in the middle and bottom panels of figure 2 as requested.

5. In the text, the authors state that high levels of fluorescent aerosols were primarily associated with flow from the NE but I don't think this statement is supported by the data presented. To me it seems that there was one period of fluorescent enhancement from the NE and one from the W. There are possibly even two instances of high loadings with westerly flow if you consider the noisy but relatively elevated concentrations at the beginning of the project in addition to what was seen in the 5th highlighted period. Other instances of flow from either direction don't necessarily bring

elevated concentrations and I don't know what the explanation is for this behavior but I don't think it's as clean as currently presented.

We will rephrase this to state that while there are both wind events featuring high fluorescent concentrations from the NE and SW, only the NE wind event A features any significant PBAP cluster concentrations.

6. The authors also state that they see enhancements in the ratio of fluorescent to total aerosol at particular times. It is nearly impossible to assess this ratio from the graph presented. I recommend adding a panel or a figure to show a time series of the fluorescent fraction, possibly showing two traces where one shows the "dust-like" fluorescent fraction and one shows the PBAP fluorescent fraction.

We agree that is difficult to determine the fluorescent and PBAP to total aerosol concentration ratio from the figures presented in the manuscript. Showing the ratio time series as a panel in figure 2 made the figure too busy so it was omitted. We feel the best way to show the ratios are as a polar plot to demonstrate the influence of wind speed and direction, which we provide below and will include in the revised manuscript.

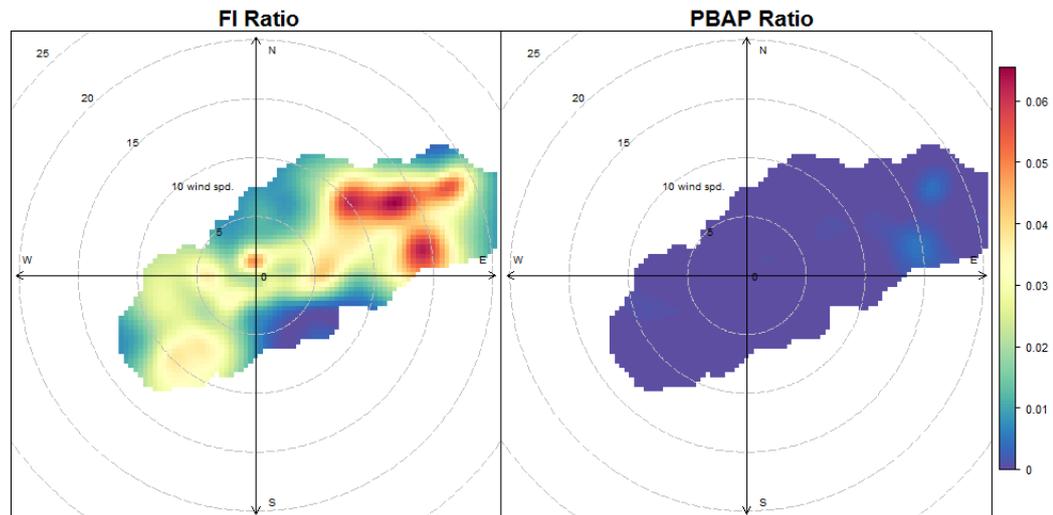


Figure 2. Polar plot of the ratio of fluorescent (left panel) and PBAP (right panel) to total aerosol concentration. Polar plots are a function of wind speed and wind direction, with concentric rings representing  $5 \text{ ms}^{-1}$  increments.

7. I don't fully understand Figure 3. Was this made from the average of all periods when the wind was from the NE and, if so, how was this average calculated? How is it that the plot for total fluorescent particles has a component in the SW quadrant but the other two do not? In panel b, it is labelled as dust but also as Cl1. I thought dust was Cl3 and Cl1 was unclassified. Either way, why show the plot for one but not the other?

The presented figures are for the time period specified for wind event A to examine the influence of wind speed and direction, demonstrating a "hot spot" ENE at wind speeds  $> 10 \text{ ms}^{-1}$ . We accept that the period being examined is not clear and we will clarify this in the revised manuscript. We will also revise the labelling of Cl1 to unclassified in the text to be consistent with table 1. We chose to show Cl1 due to its similar wind response to PBAP.

8. In the caption of Fig 4 it is stated that these plots are only for the NE wind event with the highest fluorescent loadings however the text on lines 1-3 of the same page implies that it is for all of the selected events. Please make these consistent. If the graphs are really only for a single event, it would be interesting to know whether similar behaviour was observed during other periods. What does it look like if similar graphs are made for the westerly event that had relatively high fluorescent loadings?

The figure caption displays this correct period (wind event A). We will correct the text to state this.

The SW wind event (E) does display an increase in the total fluorescent concentration with increasing wind speed, however, very little of Cl1 is observed and virtually no PBAP. The fluorescent ratio is also constant with increasing wind speed during this event.

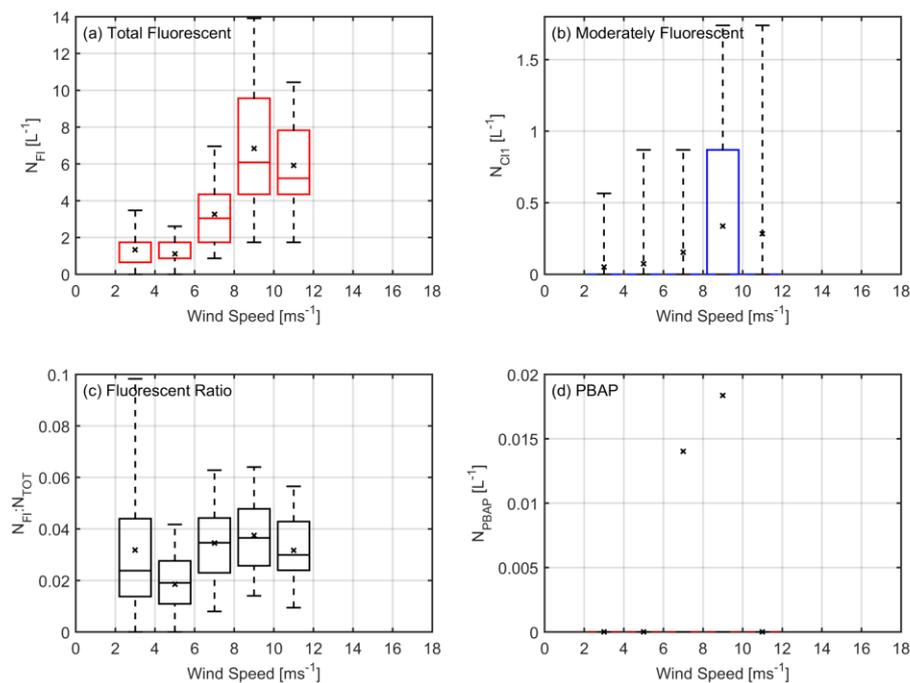


Figure 3. Fluorescent particle concentrations as a function of wind speed for the period 14/12/2015 - 16/12/2015 for: (a) Total fluorescent particles,  $N_{FL}$ ; (b) Moderately fluorescent particles,  $N_{Cl1}$ ; (c) Ratio of total fluorescent particles to total particle concentration  $N_{FL}:N_{TOT}$  and (d)  $N_{PBAP}(N_{Cl2}+N_{Cl4})$

9. I am not well-versed in calculations of flux, and I cannot speak to the validity of the method used here. In any case, I don't really see the point of calculating a flux under the present circumstances. If the elevated concentrations are episodic and not systematically associated with a particular flow direction or meteorological context, then it doesn't seem that this is likely to represent flux from sea ice or the ocean or any other dispersed source but, rather, will represent flux from a particular but unidentified bioaerosol source at an unknown location and I don't see the utility. Flux from the local environment might be better assessed by looking at wind events without elevated fluorescent concentrations but, again, I don't know enough about flux to know if this would be robust or even possible.

Previous studies have used simple concentration enhancements as a function of wind speed to imply local emissions and emission fluxes from surfaces (e.g., Sesartic and Dallafior, 2011, and references therein). We show that such approaches are overly simplistic and more robust micrometeorological methods will be needed for bioparticle flux estimates, particularly in these ice dominated ecosystems.

10. With regard to the air mass trajectory analysis, it would be nice to see maps for all of the events discussed. Was event A the only time that flow arrived from S. America or was there a time with similar back trajectories but little fluorescent aerosol enhancement?

We selected the air mass trajectories to display events of interest for comparison. We will amend figure 7 to include a representative trajectory from each event. There was only one other period displaying significant flow from S. America (27/11/15, shown below), however, this coincided with some of the lowest fluorescent concentrations observed, with no PBAP detected.

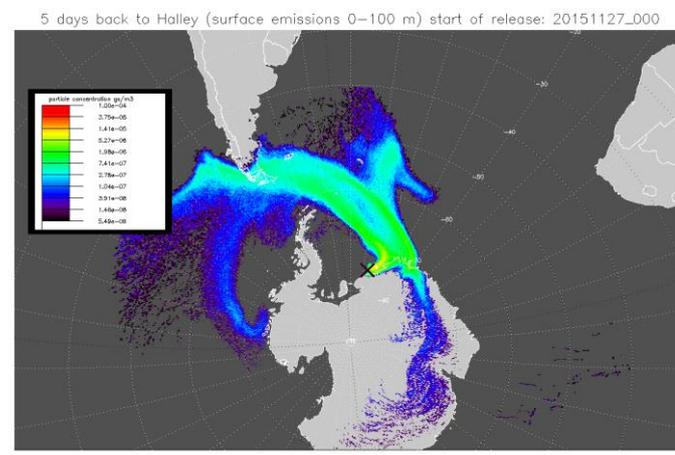


Figure 4. 5-day back trajectory analysis using the NAME particle dispersion model for 27/11/15.

11. As stated above, much of my discomfort with this paper arises from the fact that so much of the discussion centers on analysis of 5 events (and of those 5, only one or two get much attention) and the selection of these events is unclear to me. It is therefore difficult to develop a sense for how representative they might be, how to interpret the variability between them or what they mean in a larger context. The text is often written as though systematic relationships have been found which I find a bit misleading given that the study duration was relatively short and these “relationships” are extrapolated from single events. I recommend rephrasing these statements and revisiting the data analysis to more clearly delineate the observations themselves, the generalizations made based on the observations and the limitations to these generalizations imposed by the short duration of the study and the episodic nature of the environment.

It is not our intention to present the measurements from this short, opportunistic pilot study to be generally representative of Antarctic bioaerosol. As we replied to referee #2, we will reiterate the case study nature of the work presented in the final paragraph of the manuscript and we will suggest that further long term studies with accompanying supporting measurements are needed to build up a climatology of bioaerosol events to assess the influence of long range transport of PBAP/pollen from South America.

## References

Kaye, P. H., Aptowicz, K., Chang, R. K., Foot, V., and Videen, G.: Angularly Resolved Elastic Scattering from Airborne Particles, *Opt. Biol. Part.*, 31–61, 2007.

Renfrew, I. A. and Anderson, P. S. (2002), The surface climatology of an ordinary katabatic wind regime in Coats Land, Antarctica. *Tellus A*, 54: 463–484. doi:10.1034/j.1600-0870.2002.201397.x

Ruske, S., Topping, D. O., Foot, V. E., Kaye, P. H., Stanley, W. R., Crawford, I., Morse, A. P., and Gallagher, M. W.: Evaluation of machine learning algorithms for classification of primary biological aerosol using a new UV-LIF spectrometer, *Atmos. Meas. Tech.*, 10, 695-708, <https://doi.org/10.5194/amt-10-695-2017>, 2017.

Savage, N., Krentz, C., Könemann, T., Han, T. T., Mainelis, G., Pöhlker, C., and Huffman, J. A.: Systematic Characterization and Fluorescence Threshold Strategies for the Wideband Integrated Bioaerosol Sensor (WIBS) Using Size-Resolved Biological and Interfering Particles, *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2017-170>, in review, 2017.

Sesartic, A. and Dallafior, T. N.: Global fungal spore emissions, review and synthesis of literature data, *Biogeosciences*, 8, 1181-1192, <https://doi.org/10.5194/bg-8-1181-2011>, 2011.

van Lipzig, N. P. M., Turner, J., Colwell, S. R. and van Den Broeke, M. R. (2004), The near-surface wind field over the Antarctic continent. *Int. J. Climatol.*, 24: 1973–1982. doi:10.1002/joc.1090