

For item 1, I think it would be appropriate to add a brief discussion about the potential for non-biological particles to be labeled as fluorescent even with a 9-sigma threshold and your reasoning for why that is unlikely to be a problem here.

We have added a paragraph to the end of Section 4.2 explaining why soot and other non-biological particles are not thought to be influencing our observations. This is written below.

Although the 9 sigma threshold we have used should eliminate weakly fluorescent non-biological particles, the potential for more highly fluorescent particles to act as interferants should be discussed. Soot is one example, with previous studies having observed higher fluorescence than is typically seen for non-biological particles. Despite this, there are multiple reasons that we do not believe interferants are contributing to particle concentrations. Firstly, studies that found soot to fluoresce above their thresholds had typically only done so when using 3 sigma thresholding. Toprak and Schnaiter, (2013) found propane flame soot to only weakly fluoresce in FI1 at this threshold, and so we would not expect it to be considered fluorescent at a more conservative 9 sigma thresholding. Secondly, the size of the observed fluorescent particles are larger than we would expect for soot. Toprak and Schnaiter, (2013) found generated soot to only be 0.8 μm after significant coagulation time in the NAUA chamber, while Savage et al., (2017) used a mechanically dispersed dry diesel soot powder to investigate potential interferent aerosol fluorescence. They noted that this powder fluoresced above a conservative 9 sigma threshold, but this sample aerosol was much larger than soot typically observed in the atmosphere when aerosolised ($\sim 1.1 \mu\text{m}$). Savage et al., (2017) also acknowledged that fluorescent intensity is a strong function of particle size owing to surface area/volume effects and that this test soot was likely to be significantly more fluorescent than ambient diesel soot as a result. Furthermore, Savage and Huffman et al., (2018) acknowledge that more highly fluorescent soot is representative of freshly generated soot close to source, and is not representative of aged or processed soot. Ambient soot at CVAO should not be fluorescent at 9 sigma. While it is possible that soot could have internally mixed with dust and therefore become larger, this would still represent aged soot and would be less fluorescent.

I would also recommend adding text about point 2, regarding the potential effect of using data from two different models of WIBS instruments

In Section 2.1.1 we have inserted some text discussing the use of two models of WIBS.

Calibration of the LAAP-ToF was performed with pure hematite samples (Liu et al., 2018), whilst both the WIBS-4A and WIBS-4M were calibrated using NIST latex calibration beads and fluorescent glass beads, e.g. Crawford et al. (2015). It should be emphasised here that the WIBS-4A and 4M are almost identical instruments, with the only differences being the trigger levels and flow rates used. A detailed description of the 4A can be found in Savage et al. (2017) and of the 4M in Forde et al. (2019). As such, the fluorescence data for each instrument are comparable, although we acknowledge there can be issues even when comparing measurements from two identical models. An inter-comparison of the fluorescent responses between instruments when using the NIST calibration particles help affirm the instrument's similarities. This is the same methodology as has been described by Forde et al. (2019), Savage et al. (2017) and Crawford et al. (2014).

With regard to item 4 (the issue of the non-specificity of the mass spec markers for biological particles), I can see your point about how the higher threshold, based on the Savage paper, should do a better job of rejecting non-biological things. However, the fraction of particles that you are identifying as biological is very small (generally <1%) and I'm not sure that the graphs in Savage et al., are fully conclusive at that level. Even if HULIS is mostly rejected at the 9-sigma threshold, it seems that just a few misidentified particles could swing your numbers quite a bit and that cluster is just barely above the threshold. Did you check whether those mass spec markers were correlated with dust or biomass emissions? If they are that might indicate that a tiny fraction is bleeding through even at the higher threshold. At a minimum I think a little bit of discussion is warranted.

When dealing with such small percentages we agree that even small errors in classification can produce significant swings in concentrations. We believe the close correlation with particle counts for the LAAP-ToF's 'bio-silicate' class offers the strongest evidence that bleeding is not occurring, as interferant particles would have a different mass spectral profile. The role of interferants can be further discounted when considering other particle properties, for example the larger size of our biological fraction. A paragraph discussing this has been added to the end of Section 4.2.

We also acknowledge the fraction identified as biological is small (<1%) and that concentrations would consequently be significantly affected by even minor errors in the classification of particle types. However, if a fraction of non-biological particles were 'bleeding' through and influencing our concentrations, their mass spectral signatures would differ from our 'bio-silicate' class. As there is a close correlation between the bio-silicate particle counts and our fluorescent fraction, we do not believe that bleeding is significantly changing our observations. More studies comparing such a technique may elucidate the degree to which bleeding occurs, but we believe our study provides a good first estimate of bioaerosol concentrations in this region. As discussed by Savage et al., (2017), UV-LIF results should be considered uniquely in all situations with appreciation of possible influences. We are confident that many common interferant particles such as soot can be further discounted when evaluating properties such as particle size, as well as an appreciation for modelled back trajectories and identified source regions.