Response to Anonymous Referee #2

We thank the reviewer for the constructive suggestions/comments. Below we provide a point-by-point response to individual comments (reviewer comments in italics, responses in plain font; page numbers refer to the ACPD version; figures used in the response are labeled as Fig. R1, Fig. R2,...).

Comments and suggestions:

To my experience, their fluorescent # concentration was too high for Nanjing. Based on previous UV-APS data, it was about 104/m3 in the summer (June-July time period). It was also observed that there were three fluorescent peaks (1, 2.5, and 3 um). Maybe the results were different because of different instrument and different time of the measurements. In Nanjing, probably fungal spore concentration levels are higher. If they can provide some culturable or PCR data, it would significantly improve their paper.

Responses and Revisions:

A previous study on bioaerosols in Nanjing (Wei et al., 2015) was performed by means of UV-APS (excitation at 355 nm and emission at 450-575 nm), which is similar to the FL3 channel of WIBS. Although Healy et al. (2014) found strong correlation ($R^2=0.78$) between FL3 channel and UV-APS, there was a systematic overestimation of the number concentration (~3 times), which is likely to be the different threshold selection method applied. In our study, the threshold for FL3 channel is set as 18 a.u., which is calculated based on equation (1). However, comparable number concentrations with clean environments ($\sim 0.1 \text{ cm}^{-3}$; Gabey et al., 2010; Huffman et al., 2012) and in the previous Nanjing study (~0.04 cm⁻³; Wei et al., 2015) can be achieved by setting the threshold to 80-200 a.u. (FL3), as shown in Fig.R1. The impact of the applied threshold level might be more critical in polluted areas than the clean environments due to the higher fraction of anthropogenic emissions. Hence, the requirement of establishing standard calibration procedures is required in future studies. As mentioned by the referee, different sampling times might also introduce a bias in number concentrations. We have clarified this in the revised manuscript.



Figure R1. Average number concentrations of FAPs in FL3 channel using different threshold. The shaded area indicates the previous measured number concentrations in clean environments ($\sim 0.1 \text{ cm}^{-3}$; Gabey et al., 2010; Huffman et al., 2012) and in the previous Nanjing study ($\sim 0.04 \text{ cm}^{-3}$; Wei et al., 2015).

The SORPES station was influenced by the anthropogenic activities (Herrmann et al., 2014), indicating that local fluorescent aerosol particles may contain some non-biological fluorophores. We agree with the referee that comparison with culturable or PCR data would improve any WIBS data analysis and characterization of bio-aerosol, respectively. Unfortunately, this kind of information was not gathered during the campaign and cannot be retrieved in retrospect."

Comments and suggestions:

It was still not clear that how much percent of the measured fluorescent particles can be attributed to real microbial aerosol particles (e.g., bacteria and fungal spores). The authors mentioned that combustion generated aerosols might contribute to the fluorescent particles. Could the authors further expand the discussion about the types of combustions? e.g., agriculture burning, traffic, cooking, coal burning and etc. What was the major contributor?

Responses and Revisions:

In our study, we found that biological particles cannot be explicitly identified by WIBS, especially in the polluted environment. Therefore, we proposed two alternative data retrieval methods as proxy to distinguish bioaerosols. Two groups were classified as "non-combustion-related", tentatively bioaerosols type particles, accounting for ~15% and ~16% to the total fluorescent aerosol particles. Still, these two methods can't distinguish specific species. As a result of our study, and in agreement with the referee's suggestions, we propose to complement WIBS observations with other techniques, such as molecular techniques (PCR, sequencing methods, and so on) in future studies in highly polluted environments.

Figure R2 shows the particle number size distributions from different sources. Our measurements revealed a pattern in the coarse mode ($D > 1 \mu m$) which is similar to biomass burning particles (Hungershoefer et al., 2008). Both of them are dominated by the particles in the size range of 1-2 μm , with the fractions of 90% and 80% of the total number concentrations, respectively. On the contrary, particles from diesel vehicle emissions (Morawska et al., 1998) showed a different distribution in the coarse mode, and the contribution of particles in the size range of 1-2 μm was only 37%. Pöhlker et al. (2012) reported that interferents like PAHs, can be particularly enriched on the surface of soot particles from biomass burning. Therefore a possible origin of these CR particles might be biomass burning.



Figure R2. Comparisons of particle number size distributions from different sources.

Comments and suggestions:

Their results are based on one city measurement, and some results might be limited because of different climatic conditions, ecology settings and human activities.

Nonetheless, it seems the diurnal pattern was similar to those of other parts of China and the world because of the boundary layer effect. In future studies, it would be great to see the fluorescent particle diurnal pattern for those regions without boundary layer effect or at least minimal.

Responses and Revisions:

As referee mentioned, the effect of the boundary layer may result in the similar diurnal variation of different kinds of fluorescent particles. To minimize the impacts of transport and boundary layer, we also demonstrated the diurnal variation of fluorescent particles fraction, and the different diurnal pattern was found for various fluorescent aerosol particles, as shown in Fig. R3.



Figure R3. Diurnal variations of number concentrations of (a) type A, red, (b) type B, blue, (c) type C, dark yellow, (d) type AB, green, (e) type BC, purple and (f) type ABC, light blue. Gray line indicates the number fraction of respective total fluorescent particles (right axis). Shading indicates \pm one standard deviation.

The following two approaches can be used to untangle the boundary layer effect: (1) using modelling tools to account for the boundary layer effect and investigate the concentration diurnal profile under different emission schemes; or (2) performing aircraft measurements (or other high altitude) above the boundary layers.

References

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