Comments on "Ambient measurements of fluorescent aerosol particles with a WIBS in the Yangtze River Delta of China: potential impacts of combustion-generated aerosol particles" by Yu et al.

This paper described a result from the ground-based measurements of ambient fluorescent aerosol particles (FAPs) using a commercial sensor, Wideband integrated bioaerosol spectrometer (WIBS). To the best of my knowledge, this work for the first time presents the highly-time-resolved variation of FAPs concentrations over China. The topics with which this paper deals meet the scope of Atmospheric Chemistry and Physics; however, there are a lot of points to be addressed before accepting the manuscript as an ACP paper. Please consider the following comments for the revision.

Major comments

1. How to report the concentrations of FAPs classified by the measured fluorescence spectral pattern

Authors basically reported the number concentrations of FAPs as FLx, where x is channel number. As shown as figures (e.g., Fig. 6), some FLx particles have a significant fluorescent intensity at a channel other than x. It is expected that the sum of FL1, FL2, and FL3 concentrations can exceed those of all FAPs (somewhat confusing). As WIBS has a function to detect wavelength-band fluorescence, the observed data sets can create automatically seven types (= 2^3 - 1) of FAPs, where there is no overlap. Perring et al. (2015) presented this approach as authors also did as a part of the results. I recommend removing the descriptions on FLx typology and rearranging the data analysis of the seven-type FAPs at the first step to interpret how the FAPs concentrations varied during the observation period. This can improve the readability of the manuscript.

2. A message in Summary

Authors suggested the presence of "some other fluorophores" through the discussion on the comparison between non-combustion related FAPs at Nanjing and FAPs observed in other different "clean background" areas. As the atmospheric environment, ecosystem, human activities, and some other factors can greatly affect the emission of bioaerosols, the concentration levels of bioaerosols can be different among places and not be necessarily same. To the best of my knowledge, no one knows the true values of bioaerosols concentrations at Nanjing. If there is no evidence to support this message, authors should remove this sentence and modify the sentence line 322-325.

3. Approach of the classification using the fluorescent intensity at channel 3

Authors only classified FL3 (type C, BC, AC, and ABC) particles into non-combustion related (NCR) and combustion-related (CR). Although type A, B, and AB particles, which consist of a large part of all FAPs, they are not included in the classification. Why did authors use only the fluorescent intensity at channel 3 (I_3)? A simple way to see the correlation coefficient between specific type FAPs and BC/PM ratios suggests that type A and AB (type B) should be categorized into CR (NCR). If authors use only I_3 information, they do not need to deploy WIBS, and simply should do UV-APS which has almost the same function. It is pity that important and useful information is not included in the data analysis presented in this paper.

I recommend as follows.

- Please explain the benefits to deploy WIBS instead of UV-APS at Nanjing in this study if you use only I_3 for the classification of FAPs.
- A large fraction of FAPs, type A, B, and AB, should be considered and included into the classification.

4. Terminology

PAHs emitted with BC through the incomplete combustion are originally in gas phase and subsequently can be scavenged by the preexisting surface of aerosol particles. Therefore, BC is one the carriers of PAHs. It is the fact that almost all of PAHs share the emission sources with BC. However, all the particles associated with PAHs cannot be combustion-generated, are just combustion-related. I recommend modifying the terminology of “combustion-generated”.

5. Interpretation

Authors analyzed in detail the size-dependence of FL3 fraction classified by I_3. To the best of my knowledge, Figure 9 is one the most important results in this study. Positive correlation of BC/PM and FL3 fraction was clear for the size range of 1-2 µm.

I have some questions on the interpretation of the results as follows.

- How did authors set the threshold value of I_3, I_{cri}? I'm confusing to see some findings in Figure 9 such as that the FL3 fraction for the size range of 4-5 µm with $I_3 > 18$ was very weakly correlated with BC/PM and that the FL3 fraction for the size range of 5-15 µm with $I_3 > 18$ (< 80) was positively but very weakly correlated with BC/PM. The former suggests the FL3 fraction for the size range of 4-5 µm with $I_3 > 18$ can include the CR particles. The latter does that the FL3 fraction for the size range of 5-15 μm with 18 < I_3 <80 can include the NCR particles. Especially, I could not understand that authors identify the FL3 particles for the size range of 5-15 μm with 18 < I_3 <80 as CR particles. Please describe or guess what such huge combustion-related particles are. If not, we, the readers of this paper, will be confused.

- In the section 3.1, authors showed the presence of CR particles which are FL2-related (type B, AB, BC, and ABC) and have the size of 4-5 μm. As the size ranges of CR particles defined in the section 3.3.2 were limited to 1-2 μm and 5-15 μm, the definition is inconsistent with the fact shown in the section 3.1. This can confuse the reader of this paper. Please recheck the assumptions and results and make the descriptions clearer.

Minor comments

1. Introduction

Line 56-57:

Some of microorganisms cannot be cultivated. Please include this factor in the Introduction.

Line 60-74:

This paragraph is lengthy. Some details of the technical specification of commercial are not necessarily included in "Introduction" and those of WIBS should be moved into the experimental section. Why did authors include only the commercial one? Some custom-made UV-LIF instruments have ever been developed in previous studies such as Pan et al. (2009; 2011), Taketani et al. (2013), and Miyakawa et al. (2015). For the purpose to introduce the previous studies, authors should include more widely the UV-LIF techniques.

2. Methods and instrumentation

2.2. Instruments:

What is the upper limit of the particle number concentrations that WIBS-4A can accurately measure? Based on OPC-like techniques, very high concentrations can affect the counting efficiency through the coincidence error. Please clarify whether WIBS-4A works well in such highly polluted region.

3. Results and discussion

3.1. General characteristics of fluorescent aerosol particles

Line 144-146:

The "ratio" approach can minimize the effects of some processes such as diurnal variations of PBL height and air mass dilution. To the best of my knowledge, this should be valid assuming no additional formation and loss process for both numerator and denominator species. Please clarify whether this assumption is valid.

3.2. Non-biological fluorescent aerosol particles

Line 184-195:

Miyakawa et al. (2015) did not use similar technique. They used a multivariate analysis of the temporal variations of number concentrations of 8 type FAPs. This sentence is very confusing. This previous study should be included in "Introduction", because the results shown there closely relate to this study.

3.3.Classification of fluorescent aerosol particles

3.3.1. Spectral patterns of fluorescent aerosol particles

Line 200-212:

Please clarify what fluorescent compound I and II are. Are they representative compound for the combustion- and non-combustion-related aerosols? Unless they are, I have an impression that authors picked up some compounds to well account for the observation results.

Line 213-230:

As noted in "Major comments", if you use only I_3 signal, the information on type A, B, and AB particles should be ignored. Please consider some modification to the approach (See the "Major comments" for details).

4. Summary

Some sentences should be modified according to the revision. The last paragraph should be removed or moved to the discussion part, because all the descriptions are speculative, not suggested solely based on this study, and should not be discussed in Summary.

Technical comments

Line 63-64:

UV-APS use the UV-laser for exciting the particles, so here UV-Laser induced fluorescence (UV-LIF) is correct.

Line 79:

Miyakawa et al. (2015) deployed a custom-made UV-LIF instrument (not UV-APS and WIBS).

Line 107:

Is the silica gel dryer TSI's one or custom-made? If this is TSI's one, particle transmission efficiency for the coarse mode particles is not so good depending the sampling flow rate. If custom made, please clarify how authors locate it in front of WIBS-4A. The direction of flow in the dryer should be parallel to the sampling line.

Line 118:

Why did authors show approximate value of the size of a PSL particle (~2 μm)? Please provide the exact sizes and type (Sample bottle has) of PSL particles given by Duke Scientific.

Line 130:

PM_{800} is confusing. We traditionally label the subscript of PM (particulate matter) based on the size cut in "micrometer". Please modify PM_{800} into $PM_{0.8}$.

Figure 10:

I feel this figure is meaningless because Tables 2, 3, and 4 covers what this figure illustrates.

References for the comments

Miyakawa, T., Y. Kanaya, F. Taketani, M. Tabaru, N. Sugimoto, Y. Ozawa, and N. Takegawa (2015), Ground-based measurement of fluorescent aerosol particles in Tokyo in the spring of 2013: Potential impacts of nonbiological materials on autofluorescence measurements of airborne particles, J. Geophys. Res. Atmos., 120, doi:10.1002/2014JD022189.

Pan, Y. L., R. G. Pinnick, S. C. Hill, and R. K. Chang (2009), Particle-fluorescence spectrometer for real-time single-particle measurements of atmospheric organic carbon and biological aerosol, Environ. Sci. Technol., 43(2), 429–434.

Pan, Y. L., S. C. Hill, R. G. Pinnick, H. Huang, J. R. Bottiger, and R. K. Chang (2010), Fluorescence spectra of atmospheric aerosol particles measured using one or two excitation wavelength: Comparison of classification schemes employing different

emission and scattering results, Opt. Express, 18(12), 12,436–12,457.
Pan, Y. L., S. C. Hill, R. G. Pinnick, J. M. House, R. C. Flagan, and R. K. Chang (2011), Dual-excitation-wavelength fluorescence spectra and elastic scattering for differentiation of single airborne pollen and fungal particles, Atmos. Environ., 45(8), 1555–1563.
Perring, A. E., Schwarz, J. P., Baumgardner, D., Hernandez, M. T., Spracklen, D. V., Heald, C. L., Gao, R. S., Kok, G., McMeeking, G. R., McQuaid, J. B., and Fahey, D. W. (2015), Airborne observations of regional variation in fluorescent aerosol across the United States, J Geophys Res Atmos, 120, 1153-1170, 10.1002/2014JD022495.
Taketani, F., Y. Kanaya, T. Nakamura, K. Koizumi, N. Moteki, and N. Takegawa (2013), Measurement of fluorescence spectra from atmospheric single submicron particle using laser-induced fluorescence technique, J. Aerosol Sci., 58, 1–8.