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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigation of coastal sea-fog formation using the WIBS (Wideband Integrated Bioaerosol Sensor) technique" *by* Shane M. Daly et al.

Anonymous Referee #2

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Daly et al. submitted a manuscript for review titled "Investigation of coastal sea-fog formation using the WIBS (Wideband Integrated Bioaerosol Sensor) technique." The manuscript observes and analyzes data from a WIBS-4 that was deployed in Haulbowline Island, Cork Harbor during July and September 2011. The author states that size and fluorescence profiles indicated that the origin of the signals were not biological in nature. A second single-particle fluorescence spectrometer, a WIBS-4A system, was used for complementary laboratory studies to help explain field results. The laboratories studies are thought to explain a possible mechanism seen by the WIBS-4 system deployed in the Cork Harbor, which suggests the idea of the adsorption of molecular iodine onto water droplets to form I2(H2O)x complexes. The study elutes to an un-

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suspected stabilizing transport mechanism for iodine in the marine environment and provides the first real-time link between molecular iodine release, particle formation and sea-fog formation. In general, I support the publication of this manuscript in some form, however I believe there are several comments that need to be addressed before consideration. There are several peer-reviewed publications that have explored different analysis strategies for the WIBS that were not mentioned, nonetheless should be considered and discussed. I list some suggestions for specific additions below, including some possibilities for added discussion and some suggestions.

General comments:

Section 2 Methodology: In general, I think Section 2 needs a more detail on A) the laboratory experiments that took place using the WIBS-4A instrument and B) the preparation for the data for both the field analysis and complementary laboratory studies. Since this manuscript is the first evidence of using real-time fluorescence spectrometers to observe the link between molecular iodine release, particle and sea-fog formation, I think it is crucial for the methodology to be written so that it can be repeated and further explored, specifically in Section 2.3. There has been several studies on the preparation of WIBS data, before subsequent analysis and how this may change the overall observations, e.g. Gabey et al., 2010, Perring et al., 2015, Savage et al., 2017 and Savage et al., 2018, and references there-in. All the mentioned studies look into fluorescence thresholding and how this may result in the efficiency at which the WIBS can discriminate between biological and non-biological particles. From my understanding the author uses the FT signals as the fluorescence threshold, and compares results to what was seen in the Hernandez et al., 2016 publication, however this publication uses the default FT + 3sigma threshold. I suggest the author goes into more detail on why they chose to use the threshold they did, and what implications this may have on their results. Some of this extensive detail may belong in the discussion, however how the author 'prepares' the data for both the laboratory and field study should be more explicit (e.g. size calibration information, fluorescence calibration information,

the fluorescence threshold chosen- whether it is the average, median, etc.).

Section 2.2 Field Instrumentation: On page 6 lines 21-24, the author states that both the WIBS-4 and the WIBS-4A units were identical in terms of functionally- this is strong statement. Studies including Robinson et al., 2017, Savage et al., 2017, and Tobias et al., 2018 explain the current hurdles to when comparing data from two different WIBS units (or fluorescence spectrometers). Hernandez et al., 2016 uses two different WIBS units in his studies, and it can be seen first-hand the differences in fluorescence signals produced by two different instruments observing and measuring the same particle type. The Robinson et al., 2017 publication provides a procedure for calibrating the different WIBS channels for the inter-comparison of WIBS data. Can the author please comment on whether such calibration was done? Where the PMT voltages measured for each WIBS unit?

Page 11, lines 5- 20: In general, I think this section needs more discussion in regards to the suggested publications and their analysis strategies - Gabey et al., 2010, Perring et al., 2015, Savage et al., 2017 and Savage et al., 2018. It is not clear what the author means by stating " Unusually, fluorescence signals were mainly measurable in the FL1 channel. FL2 registered little emission above threshold as illustrated in Figure 4, which shows plots of size/AF data as a function of the FL1 and FL2 channels. (FL3 showed no fluorescence). The larger size feature (2-6 um) consisting of highly fluorescent solid particles/droplets but only in the FL1 channel represents a behaviour that has not been observed previously in any WIBS field campaign. Hence fungal spores, certain pollen and bacteria as large as 2 um (Hernandez et al., 2016) can be found in the 2-6 um size regime but are fluorescent in all channels because of their amino acid, tryptophan and NAD(P)H contents". The excellent study by Hernandez et al., 2016 provided the first extensive characterization of the WIBS using various biological particles. It is important to keep in mind the many particles, e.g. pollen and fungi, are larger than 6 um in diameter and one's aerosolization technique may influence the size distribution observed. Savage et al., 2016 showed that aerosolization of pollen using turbulence

created with a stir bar resulted in fragmented pollen. On another note, the author compares fluorescence signals in this study with those observed in Hernandez et al., 2016, however this cited publication uses a different fluorescence threshold. Studies have shown that biological particles may not have FL3 fluorescence characteristics when observed by the WIBS, some of these particles include various bacteria and fungi.

Several studies suggest there are non-biological, fluorescent particles that may be interferences when discriminating between bio vs. non-biological particles, and even different particle types (Huffman et al., 2010, Pohlker et al., 2015, and Savage et al., 2017, and references there-in). Can the author comment on these possible interferences, and if these substances were taken into consideration during their field analysis?

Minor Comments: Figure 4: The y-axis legend is unclear- is it the number of particles the succeed the threshold in all three channels? Figure 3: I suggest logging the x-axis. The y access legend is unclear- see comment for Figure 4. Page 11, lines 1-4 “It is clear, from the data shown in Figure 3, that a bimodal size distribution was recorded with: (i) a highly fluorescent, broad feature observed between $\sim$2-6 μm, peaking at $\sim$2.5 μm; (ii) a much narrower peak in the size regime <1.5 μm that represents non-fluorescent particles.” Are these particles truly non-fluorescent or just ‘weakly’ fluorescent? It seems based on the gradient of the color legend in Figure 3 that most of these particles exhibit fluorescence that are indeed over the thresholds stated in page 10 line 7.

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