

Response to Referee #2 (J Alex Huffman)

*We appreciate the positive comments of Professor Huffman, and thank him for his critique. Please see responses below (in italics).*

The manuscript submitted by Twohy et al. presents measurements of fluorescent aerosol and ice nucleating particles (INPs) collected from the Gulfstream-V aircraft as well as modeled interpretations of these data. The investigation of biological particles at altitudes relevant for mixed phase clouds has been performed only a few times, but this is the first manuscript to describe the application of real-time fluorescence-based detection (i.e. WIBS here) along with analysis of INPs from an aircraft and showing vertical distributions. Overall, the manuscript is an excellent contribution to the literature and is very well written. The manuscript fits well in ACP, and I anticipate that it will be well received and well cited. I have some minor points that I think may help clarify certain aspects of the text, but otherwise I recommend the manuscript be published without major alteration.

#### Minor Comments

1) The presentation of the sites of observation and the timeline of comparison with previous studies is a bit confusing at times.

a. L82 states that Figure 1 was taken from the Southern Great Plains ARM site. I would suggest putting a mark on the map in Figure 2 to highlight this.

*Great idea. We have extended the SE quadrant of Fig. 2 so that the ARM site is now marked on the map, and mentioned in the caption.*

b. L97 discusses data “taken near Boulder, CO”. Does this refer to flight data, or ground-based measurements from the BEACHON study? If referring to the aircraft data, it is confusing, because the flight tracks extend well beyond Colorado. However, if referring to the BEACHON study, I would be much more specific.

*Boulder was near where the aircraft was based and was a convenient site to compare temperature soundings to the ARM site soundings. For clarity, we have expanded the text to: “Fluorescent biological particle measurements described later in this paper were taken farther north in Colorado, Wyoming, North Dakota and Nebraska. For comparison, seasonally-averaged surface temperatures at Boulder, Colorado are about 2K-5K colder than at the ARM site.”*

c. L139 mentioned the “BEACHON” study (in quotes), but does not list the full name. For this journal I would suggest listing the specific name as BEACHON-RoMBAS and spelling out the acronym, per convention. The first time it is mentioned, which I believe is at this point, I would also suggest referring to the site itself, which is called the Manitou Experimental Forest Observatory (MEFO), rather than the “BEACHON project site.” This may help clarify for community members familiar with the site, but not with this specific BEACHON study. An overview of the site is presented by Ortega et al., 2014.

*We have implemented both these suggestions, replacing the BEACHON study with “BEACHON-RoMBAS” and the BEACHON site with “MEFO” site, as appropriate throughout. We’ve also moved the discussion of the BEACHON-RoMBAS experiment and MEFO site down to section 3.1, with the Ortega reference, where all this information can be together.*

d. I would also suggest pointing out that the BEACHON-RoMBAS study was in July-August 2011, whereas these flights were performed in October 2013. Because the years are sometimes not reported later, it may confuse some readers who are not already familiar with these studies.

*Done in section described above and below.*

e. L241: Here is an example where I would add the year (2011) and consider changing to MEFO or adding that information here.

*Done.*

f. L304: MEFO (or the site where BEACHON-RoMBAS was performed) is near Woodland Park, CO, but not very close to Manitou Springs, CO.

*Changed to Woodland Park.*

2) The discussion of WIBS data as treated relatively carefully, but I would suggest changing the wording in a few places to make the statements somewhat more conservative. For example:  
a. L162 states that “most biological particles contain amino acids and other compounds that fluoresce ...”. True, but I would either give more detail (as in L185), or remove ‘amino acids and other’ from the sentence. As written it seems half-way between a specific statement and a vague one.

*Removed amino acids as suggested.*

b.L164, L313, L396, L423: Each of these lines give some statement implying that biological and non-biological particles can be differentiated by the WIBS. This is a nuanced discussion, as the authors mention. However, I would suggest scaling back the wording for these sections a bit to involve the word fluorescent, or some other terminology that does not inadvertently imply more knowledge than can be defended. Even though the authors do bring this up, I think it would be best to utilize terminology along the way that will help in case a reader doesn’t look carefully at the sections with these important caveats.

*Understood; we have changed the first discussion in Section 2.3 to: “Therefore the WIBS-4A may be used to distinguish fluorescent particles that are predominantly biological from non-fluorescent particles that are predominantly non-biological (Pöhlker et al., 2012; Huffman et al., 2013)”, and changed **biological particles** to **FBAP** or similar, more appropriate terminology throughout when referring to WIBS measurements. In one case (current line 217), we have changed biological to supermicron, which is more correct in this instance.*

### 3) Sizing

a. The manuscript discusses “large particles” several times, but I’m not sure they are ever rigorously defined. I think the authors use 0.8  $\mu\text{m}$  as the lower cut for “large” because of the WIBS. Please add this unambiguously when the term “large particles” is used first. I would also add an upper size range for this, since the WIBS, and probably the inlet, do a poor job of collecting very large particles.

*Good point, both to be rigorous in describing what we are presenting and also since other studies may use a slightly different size range. The lower limit is indeed due to WIBS detection limits and the effective upper limit is about 12 microns due to system transmission efficiencies. We have specified this size range now in the Abstract and added an explanation of these size limits in the second to last paragraph of Section 2: “Based on size-dependent concentration corrections for inlet aspiration and transmission efficiency described in Appendix A, net efficiency for particles larger than 12  $\mu\text{m}$  diameter was less than 2%. Detection of fluorescent particles smaller than 0.8 in diameter is limited by the sensitivity of the WIBS detectors (Gabey et al., 2010). Therefore, when presenting measured concentrations or properties of “fluorescent biological particles” or “FBAP” in this paper, only particles between 0.8  $\mu\text{m}$  and 12  $\mu\text{m}$  in diameter are represented. “*

b. See L395, but many other locations as well. c. L423

*We have now more rigorously specified the measured size range (0.8-12  $\mu\text{m}$ ) where appropriate throughout.*

4) End of page 5 discusses how the WIBS background signal was calculated. Was the “forced trigger” calculated as one average per flight, one average for all flights, a running average within a flight? Please clarify.

*We have inserted a more detailed explanation of how the forced triggers were used: “Forced trigger measurements were performed at the beginning and end of each flight, during which time the instrument fires the UV light sources in the absence of particles to measure the background signal. The background signal averages and standard deviations were linearly interpreted over each flight. Only fluorescent signals larger than the forced-trigger average value plus 2.5 standard deviations are included in the data presented here.”*

5) I would suggest adding Huffman et al. (2013) to L305 and including that droplet freezing apparatus measurements were also performed alongside CFDC measurements.  
*Now added to beginning of Section 3.1.*

6) Figure 3: Legend is a bit confusing. The caption implies that there is a difference between large and small data points, in terms of whether they pass statistical significance tests. The sizing difference is subtle, however, and I would suggest making this easier to determine from

the (i.e. shaded or not ...). Also, the legend repeats information (e.g. 6B upper bnd, 6B lower bnd), but I'm not sure if this is intentional or necessary.

*We have combined your suggestions and those of the other reviewer to combine 3b and 3c into a new 3b plot, which is larger and more legible. The legends are also modified for clarity. We have retained, however, two sets of 6A and 6B symbols in the legend, since upper and lower bounds use slightly different symbols.*

7) Figure 7: I would suggest making one legend and putting in a location of a sixth panel. This would reduce legend redundancy and would remove the current issue that the legends cover parts of the graph and axes labels.

*Done as requested.*

Specific and technical comments or corrections:

1) L 76: Comma after "Thus"

*Done.*

2) L110: Does the sentence need to be parenthetical?

*Parentheses removed.*

3) L183: Add closing parentheses.

*Done.*

4) L204: "calibration was verified" seems a bit strong for a 1-point measurement. Maybe "check" is a better word?

*Yes, changed.*

5) L278: Add space between "10hPa"

*Done.*

6) L288: Add space between "4um"

*Done.*

7) L308: Add comma after "First"

*Done.*

***Other minor corrections made to manuscript by authors:***

*We have modified the following statement: "Pratt et al. (2009) and Creamean et al. (2013) reported that biological particles sometimes **dominated** ice residuals in mid-level clouds over the western United States" to: "Pratt et al. (2009) and Creamean et al. (2013) reported that biological particles sometimes **comprised a large fraction** of ice residuals in mid-level clouds over the western United States." The prior statement was somewhat misleading, since in the Pratt paper, mineral dust actually dominated over biological particles.*

*We have modified the sentence immediately before the "Conclusions and discussion" section as follows: "**The variable and often low abundance of these INP, however, may explain why clouds sometimes remain supercooled in the atmosphere, particularly at warmer temperatures (Kanitz et al., 2011; Komurcu et al., 2014).**" This reflects new information and an important data set on supercooled clouds not known to us at the time of submission (added Komurcu reference).*