Answers to RC2 (Darrel Baumgardner)

The authors would like to thank Dr. Darrel Baumgardner (RC2) for taking his time to review the submitted manuscript, especially considering his overwhelming expertise and well-known prestige in the field of optical particle counters. We have now updated the manuscript text to address his concerns and hope that the detailed point-by-point responses presented below will convince him to reconsider his initial recommendation to withdrawal the manuscript. All line numbers in the responses refer to the revised version of the manuscript.

Comment #1: The interest of the authors in using the particle by particle data (PbP) is a worthy objective; however, they have overlooked a number of critical factors in their methodology development that puts in question the usefulness of the smoothing technique until they address these factors. Before embarking on this development and writing of this manuscript they should have contacted us at Droplet Measurement Technologies and discussed what they planned to do. This would have possibly clarified for them why their approach needed to be reviewed and modified.

Answer to comment #1: Thank you for acknowledging the effort involved in using the PbP data to eventually improve the precision in sizing with the CAS. We agree that some of the methods were unclear in the submitted manuscript. We have now taken the chance to add more complete explanations in several parts of the text that, hopefully, will make our arguments clearer.

In terms of contacting DMT, we would like to point out that we (SNV, AC and VF) had several rounds of discussions with DMT scientists and product engineers about several of the "critical factors in our methodology". DMT product engineers eventually informed us of some specific values of our instrument's functional parameters such as that of the angular interval where the scattered light is collected $(4.0^{\circ} - 13.5^{\circ})$.

Comment #2: Secondly, they have overlooked a number of important publications that have already explored the issues that they discuss and addressed how to account for the ambiguities in size and scattering cross section. I have listed these below. Although several refer to the FSSP and not the CAS, the collection angles are similar and measurement principals are the same.

Answer to comment #2: Thank you for pointing these studies out. We completely agree that we should have included them and have now done so in the revised version of the manuscript as follows: Brenguier et al. (1998), on lines 164, 194 and 415; Granados-Muñoz et al. (2016), on lines 121 and 194; Pinnick et al (1981), on lines 163 and 190; Rosenberg et al. (2012), on lines 120, 164 and 347. However, it is worth noting that the article by Rosenberg et al. (2012) was actually cited twice in the Discussion version of the paper. After looking more closely into previous approaches of the possible influence of various sizing errors on the ensuing droplet distributions, we have also included: Dye and Baumgardner (1984), on lines 120 and 190, Cooper (1981), on line 414 and Baumgardner et al. (2017), on line 139, to the manuscript.

Comment #3: Thirdly, you have to take into account two important factors when carrying out the Mie calculations: 1) the droplets are being illuminated by a laser whose intensity cross section is not precisely uniform, which means that the high resolution oscillations are smoothed out (the authors state on line 157, "In older descriptions of the forward scattering spectrometers (originally used for aerosol sizing measurements, see Baumgardner et al., 1992) this aspect seemed to be overlooked and some smoothened versions of the FWSCS vs. diameter diagrams appeared to have been used.", but we were well aware of the oscillation but took into account the multimodal aspects of the FSSP lasers. And 2) the authors need a better understanding of how the scattering angles are obtained and understand that they are not a precise 4-13.5. Why? Because the scattering angles are determined by the distance of the measured droplet from the dump spot, the diameter of the dump spot and the diameter of the aperture. This distance varies because the depth of field is of finite width. This means that the positioning of the peaks and valleys in the FWSCS shift slight, smearing out the fine detail that the authors show in their figures. This has to be taken into account.

Answer to comment #3: First of all, the authors totally agree that the phrase indicated by the Referee is inappropriate and would like to apologize for misrepresenting the study. It actually dates from the earliest of the many versions of the text and is now removed in the revised manuscript and changed to "*This aspect, which generates sizing ambiguities through FWSCS measurements, has been known and analysed for a long time in the literature (Pinnick et al., 1981; Baumgardner et al., 1992; Brenguier et al, 1998)*" on line 163.

Regarding the "Mie calculations", which we understand as obtaining the particle size distribution through a comparison with the exact FWSCS-diameter diagram, the authors

believe that there are two ways to deal with this. Firstly, the FWSCS-diameter diagram (or Mie diagram) can be constructed by embedding in it all the instrumental errors (including those resulting from the cross-sectional non-uniformity of the laser beam and the imprecise knowledge of the angular interval for collecting the scattered light). By accounting for all of these errors, the fine details of the Mie diagram will certainly be smeared out (as the referee states) to a smoother curve, which allows for the establishment of a relatively small set of uneven (and unequivocal) size bins over which the size distribution can be built up. This is the typical approach that allows fast practical evaluations of the in-flight data. A related comment has been added in the new version of the manuscript on line 189: "For practical purposes, partitions of the diameters' range in uneven size bins have been previously proposed in the literature (Pinnick et al., 1981; Dye and Baumgardner, 1984). Such procedures are actually fitting of the exact FWSCS-diameter diagram with a discrete monotonic plot of response thresholds, each corresponding to a size bin limit. The fitting should be made so that the differences between the threshold plot and the exact Mie diagram can be assimilated to the resultant of the various errors generated in the FWSCS measurement process (Brenguier et al., 1998; Granados-Muñoz et al. 2016)."

Alternatively, starting from the finest, "exact" form of the Mie diagram, one can take each measured FWSCS value and, in case it is equivocal, equally "distribute" it to all its intersections with the Mie diagram. That is to say, instead of counting one particle for one of the intersections, we count a fraction for each intersection in the size distribution. For example, if there are 5 intersections for a certain measured FWSCS value, at the diameter values d1, d2, ..., d5, consider in the distribution that, for each d1, d2, ..., d5, we have 1/5 of a particle. Then, to account for the measurement errors, which are assumed as known for each value of the FWSCS, compute the shift it produces in the distribution and the ensuing errors of other cloud parameters that result from various averages. This proposed approach, which is briefly described here, is discussed in detail within Section 4 of the manuscript. The difference between the two methods has also been outlined in two new paragraphs added in the revised version of the manuscript, starting on line 413: "At this point, it should be mentioned that the influence of the sizing errors on the resulting droplet distributions has been previously addressed in detail in the literature (Cooper, 1981; Baumgardner et al., 1992; Brenguier et al., 1998) through an ingenious mathematical method based on a transfer matrix that takes the measured distribution into the actual one. The elements of the matrix are actually probabilities that a certain measured particle of a given diameter be counted in a different size bin. The transfer matrix has to be constructed for each instrument and its elements embed both the

errors generated by the FWSCS measurements and those ensuing from the ambiguities in the comparison with the Mie diagram.

The present study tries a different approach, by separating the measurement errors of the FWSCS values (which stem from various hardware issues and have to be known) and by considering in greater detail the uncertainties generated by the comparison with the "exact" Mie diagram."

In the end, both methods basically lead to the same macroscopic result, but each one has its specific advantages. The first method is simpler, fast and suitable for rapid in-flight analyses. The second method is more computationally demanding and can be applied only post-flight and only if one knows the measured values for individual particles, which means the availability of the PbP files. As for the advantages of the second and proposed method, they all stem from the possibility of obtaining fine size distributions which may provide the cloud composition in greater detail and also could be prospectively useful in operating future, likely more precise versions of optical particle counters. Moreover, as exemplified in Figures 3 and 4 of the manuscript, the fine size distributions can be readily used to construct distributions over coarser structures of (even or uneven) size bins (one can actually reproduce – and we did this as a test of consistency – the size distributions obtained with the first method). As pointed out on line 468, coarser size distributions are more convenient for evaluating the effect of measurement errors.

Comment #4: Finally, although I would really like to see the type of fine detail in the size distributions that the authors show in Figs. 3 and 4, and attribute to natural microphysical features, I suggest that they look carefully where those peaks and valleys fall in the size distribution and then take a careful look at their FWSCS diagrams and they will see that many, if not most, of these feature are a result of the Mie ambiguities. This is why they have to read Brenguier et al who actually uses those features to do quality checking of their FSSP.

Answer to comment #4: The Referee suggests that the features appearing in the size distributions result from the Mie ambiguities. The authors cannot agree with this statement. The ambiguities of the Mie diagram cannot play a role in the appearance of various features in the size distribution constructed through the proposed methodology for the following straightforward reason: when a particle is detected with an ambiguous FWSCS value, the counter does not record several particles but several "fractions" of a single particle (as discussed on lines 250-255). So, only if there are more particles with some FWSCS value, there

will be a peak in the distribution in the size range where the given FWSCS value intersects the Mie diagram. If there are few or no particles with that value of the FWSCS, then there will be a minimum in the distribution. If the maxima or minima of the detailed size distributions would be "a result the Mie ambiguities" then all the detailed size distributions obtained through the proposed methodology should have the same appearance. However, it is sufficient to look, for example, to panels (a) and (c) of Figure 12 to observe that the detailed size distributions constructed for two flight lines show pretty different features, even if they were obtained with the same ("exact") Mie diagram.

Comment #5: Line1 67: The scattering cross section is not quasi-monotonic. It oscillates.

Answer to comment #5: The expression "quasi-monotonic" has been replaced by "oscillatory" on line 69 in the present revised version of the manuscript.

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