First I would like to state that I think that it is fantastic to see a paper like this submitted. It is of course obvious that high quality instrument calibration is key to high quality data output and papers such as these highlight such consideration to other users and to manufacturers.

Up front I would like to highlight the fact that I am first author of the Rosenberg et al. (2012) paper referenced in the manuscript which also details calibration practices used for a PCASP and a CDP (and which have also been applied to a SID2h). I therefore (rightly or wrongly) clearly have my own perspective on OPC calibration. I will refer to this paper as R2012 in the rest of this comment.

Of particular note in this paper is the assessment of the impact of nonsphericity on the response of the PCASP. It is an issue of concern to many OPC users that the size presented by an OPC is difficult to equate to a geometric metric for non-spherical particles. Any advances towards resolving these concerns are likely to be welcomes by the community, especially if the work is capable of showing whether Mie theory can adequately represent the scattering seen by the instrument from nonspherical particles. This is due to the necessity to make refractive index corrections to the bin sizes when the composition of particles used for calibration does not match that of real world particles measured in the field.

As these methods have been used for some years and are likely to have improved data quality for a number of campaigns it is useful to document them. The methods presented in R2012 differ significantly from the methods presented here. The R2012 methods were generated to be as general as possible so that they could be applied to any OPC over any size range. I feel that R2012 represents best possible practice for OPC calibration. However, in this manuscript only the submicron region of a PCASP is considered and as the response of this instrument in this size range is monotonic, simpler methods may be able to achieve good result s. With this in mind presented below are some areas where it is felt that the manuscript can be improved.

1) The methods described here differ greatly from the methods discussed in R2012. In R2012 the instrument is calibrated in terms of the particle scattering cross section, because the instrument responds linearly to this property. Here the instrument is calibrated in terms of particle diameter. There are two potential issues here
    a. The instrument response to diameter is nonlinear and for particles larger that 1 µm is non-monotonic, see Fig C1.
    b. The authors use 1 calibration point to apply an offset to the manufactures specification.

    As can be seen in Fig. C1 if the particles used happen to fall on a spike or trough on the curve they cans significantly bias the results. In this case the calibration particles used are all of diameters less than 1 micron, where the response is not so spiky. From 0.3 – 1.0 µm one cold even consider it close to linear. If this is by design then the authors should report so and comment on the uncertainty based on extrapolation. It should be noted that PSLs are available over the whole range of the instrument even if the SMPS range is limited.

    Even if the response to diameter was linear it is not clear that an offset is the appropriate correction. There are potential sources of offset in the instrument, however there are sources of sensitivity variation as well. For example, the location of the sample in the laser beam and dirtying of the optics would change the instrument response by a common

multiplicative factor. Given the points a and b above it would be good to see at least two data points per gain stage for calibration regions less than 1 micron – the minimum for a linear fit – and preferably more. This would at least allow an offset and a sensitivity correction to be derived. It would also be good to see a comparison with the R2012 methods to see if the results differ significantly.

2) If the diameters of the bin boundaries increase we may expect to see a change in the bin widths also due to the nonlinear instrument response. The authors do not attempt to calibrate bin widths and should comment on the uncertainties expected.
3) As mentioned in the manuscript, custom threshold tables can be provided to the PCASP. This was used in R2012 to zoom in on the region of interest during a calibration and could be utilised here where the accuracy is currently impacted by the standard resolution.
4) The concentrations reported for use during calibration seem rather high (10^4-10^5 cm^{-3}) At these concentrations we may expect coincidence to occur in the PCASP (i.e. two separate particles passing through the sample volume of the laser at the same time, distinct from an aggregate). Such coincidences may be responsible for some of the long tails observed which were not seen in data in R2012, see Fig C2. The PCASP reports the transit time of the particles through the laser beam meaning it should be relatively trivial for the authors to calculate the fraction of time during which particles are in the laser beam and hence the probability of coincidence.
5) The report of R2012 on p4131 line 25 onwards of the manuscript is not quite correct. We found a result that seemed consistent with particles not transferring from one gain stage to the next when it appeared that they should. We did not measure pulse heights so it is not clear if the pulse heights were lower than expected or if there existed a fault or design flaw in the PCASP hardware or firmware that caused the problem. Regarding the solution provided on p4132 line 9 onwards, we feel that this is a sensible approach and the authors should highlight that this creates a single bin which spans the gain boundary (it is not currently immediately obvious). It is somewhat analogous to the merging method of R2012, but with the advantage of maintaining the 30 bin resolution.
6) Regarding the baseline reference voltage, it is not clear to the reader whether this changes the minimum limit of the gain stage, the sensitivity of the gain stage or causes a constant offset to all bins of the gain stage. A figure showing a pulse or a series of pulses (either schematic or based on oscilloscope measurements from the instrument test points) would be useful to explain this with the baseline voltage and the equivalent thresholds marked on the y axis and would facilitate advances beyond the reporting by R2012. It is still not clear to me how exactly the baselines interact, nor does it seem that the manufacturer, DMT, is able to provide much insight. It seems that this is something that will only be established through some detailed experimental work.
7) Again when discussing Fig. 5 of the manuscript on p 4134 it would be good to know the concentrations recorded from the two measurements to understand coincidence effects as such broad tails are not seen in Fig. C2 reproduced from R2012. These differences should be explained and if they do not arise from coincidence then they could be due to the different model and operation of nebulisers, i.e. if a nebuliser produces larger droplets then it will give a higher number of aggregates.

8) What type of diameter equivalence is reported in Sect 3.7? It appears to be volume equivalence but it isn't stated explicitly. If there are coincidence effects in the data as well as aggregate effects do these impact the conclusions?
9) When investigating the scattering from aggregates the authors may again benefit from zooming in on the regions of interest to improve resolution. This may be particularly interesting for the three-particle aggregates where the authors discuss differences between linear and compact configurations.
10) There is no mention of uncertainties in the manuscript. This is important, because unless uncertainties are provided there is no indication of the value of the calibration. Given the extrapolation based on one point per gain stage it is particularly important that the effect of this extrapolation upon uncertainty is assessed.

In summary, in the region where instrument respose is close to linear the methods presented in this paper have the potential to improve data quality beyond using the manufacturer's specification. However use of only one data point per gain stage is of concern, particularly when extrapolated to the nonlinear regime above 1 μm. The impact of this over the entire size range should be assessed and if necessary the use of multiple sizes per gain stage should be adopted. The validity of a linear method for particles greater than 1 μm should be assessed. In addition the impact of coincidence should be considered upon the measurements of aggregates and this data should be highlighted in the paper as it is a significant output. It would be interesting to see if aggregates could be produced with diameters greater than 1 μm, well out of the Rayleigh scattering regime to see if equivalent volumes spheres are still appropriate, however, there may be experimental limitations which preclude this.

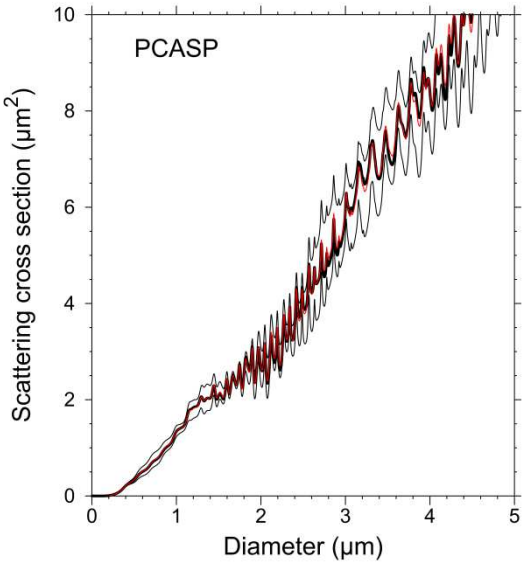

Fig C1. Scattering cross section as a function of particle diameter for the scattering angles of a PCASP, reproduced from Rosenberg et al. (2012). The scattering cross section equivalent to PCASP response as these parameters are linearly related. The bold line represents the nominal optical geomety, the fine black lines represents the situation if the sample was offset along the axis of symettry of the optics and the fine red lines represent the situation if the sample was offset perpendicular to the axis of symettry.

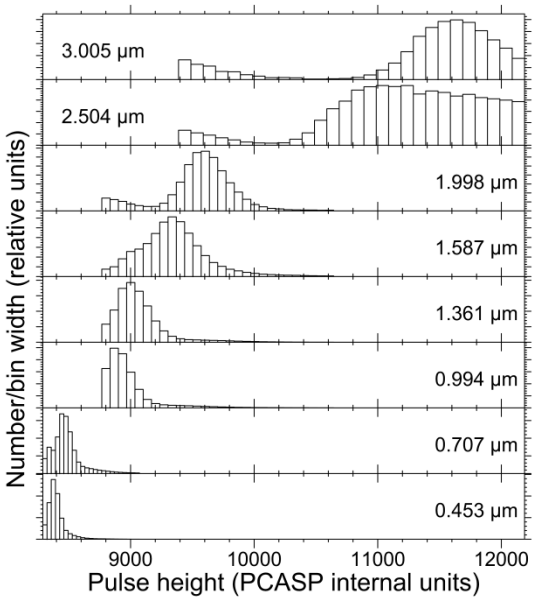

Fig. C2 Particle distributions seen for PSL beads for a PCASP. Reproduced from R2012. The PCASP was provided custom threshold values to zoom in on the region of interest. Comparison with the manuscript's Table 2 shows that for example the 0.453 µm beads would usually fall almost entirely in bins 15 and 16.