This manuscript describes an approach for interpreting measurements made with a PCASP and CDP, primarily focusing on calibration and adjustments for varying indices of refraction. This study represents a useful contribution to the general literature on the use of single particle light scattering instruments (OPCs) for atmospheric studies but should be expanded a bit more to be of further value to the community of those who use OPCs in their research.

Given that only a small number of researchers actually work directly with the instruments, i.e. calibrating and working with them “hands-on”, although the description of how to do an instrument calibration is useful, and helps those who use the data get a “feel” for how the instrument operates, of far greater value (in this reviewer’s opinion), is to give those who use the data more tools for estimating the uncertainties.

In this regard, I think that a large fraction of Section 2.1 should be removed as it is mostly a tutorial on DMAs and Mie theory that has been thoroughly described in the literature. Likewise, the description of how to measure the response of an OPC is probably more detailed than necessary, but I won’t quibble about its length and detail.

There are, however, a number of details that should be further clarified in text. First of all, the determination of the electronic response to scattering intensity is quite sensitive to the fidelity of the calculated scattering cross sections. As pointed out by the authors, this is dependent on the assumption of collection angles used in the calculations. For the CDP, Droplet Measurement Technologies no longer assumes a nominal forward scattering collection angle of 4°-12°, based on the physical geometry of the instrument. These angles are very sensitive to the distance of the center of focus from the dump spot, the diameter of the dump spot and the opening in the receiving arm. These three dimensions are determined by the optical and mechanical components that are controlled as well as possible during the manufacturing and assembly of each individual CDP but zero tolerances are not possible, hence small differences lead to small differences in the collection angles. Although these differences are small, they do lead to differences in the subsequently calculated scattering calculated cross sections – as the authors point out. For this reason, DMT now deduces the correct collection angles, by measuring seven different sizes of calibration beads of known refractive index and then runs the Mie code, iteratively changing the lower and upper scattering angles, until the calculated scattering cross section matches the seven calibration points with the smallest error. Using this approach the lower scattering angles have been observed to range between 3.3 and 4 and the upper angles from 1.5 to 13.8.

DMT has not yet implemented this approach with the PCASP but is in the process of doing doing so. This approach could be easily implemented with the calibration technique suggested in this paper using either multiple PSLs or with the scanning DMA.

The second detail that is not explained in the text is that both the PCASP and the CDP use polarized, Gaussian mode lasers. This means that the Mie calculations have to incorporate polarization vectors and, even more importantly, the users of the data from these instruments must understand that the nature of the Gaussian mode is such that not all particles of the same size will intercept the beam at its point of maximum intensity. Indeed, in the PCASP the aerodynamic jet is designed to keep the majority of the particles in the center, most intense portion of the beam, but there will always be some portion that pass through less intense portions and are hence undersized and leads to broadening of the distribution.

In the CDP, the rectangular mask on the qualifying detector is designed to accept particles that pass only within the most intense portion of the beam but the intensity pattern is still Gaussian and there will be a spread in the scattered intensities for the same particle size.

This spreading of the size distribution is no different than the spreading seen in the DMA where elaborate transfer functions have been devised to invert the data from an SMPS to derive the size distribution. There is no reason that a similar transfer function couldn't be devised, as I will elaborate on below.

Measurement Uncertainties related to refractive index

The authors suggest that the best methodology for processing the size distributions is to set up the size thresholds for the channels of the specific instrument using probabilities that given an assumed refractive index, the particle of size D will fall in one of the size bins defined for the instrument. It was not at all clear to me how these probabilities were calculated so I went about doing this for myself to get an idea how this would work.

For example, assuming that the CDP is correctly set up so that a 50 μm water droplet, refractive index = 1.33, will fall in the top bin with an A/D count of 4095 (as shown in the threshold table for the nstrument). I can calculate a scale factor by calculating the scattering cross section, I_c, for a 50 μm droplet whose scattered light is collected over the forward scattering range of 4-12°. Then each of the 30 channel thresholds can be converted from counts, C, to scattering cross section, I, by multiplying by $I_c/4095$.

The probability that a particle with diameter D, will fall within any of the 30 channels is then derived by assuming a specific refractive index, calculating its cross section and finding which channel it would fall in.

For example, using a refractive 1.33, as shown for the first 10 channels of the CDP in Table I, we find that 58% of the particles in Channel 0 come from particles with this size (2-3 μm) while 42% come from particles whose size should have put them in Channel 1 (3-4 μm) but are instead put in Channel 0 because of the oscillating nature of the Mie curve. Likewise only 9% of particles in Channel 1 actually are coming from particles that should be in this been since many have been sized in Channel 0 while the remainder come from the three larger bins above (32, 32 and 25%, respectively).

This same exercise can be repeated but with mixtures of particles with differing refractive indices. For example, Table 2 is a mixture of 6 refractive indices and Table 3 a mixture of 12, including absorbing particles.

Table I
Probability Table for refractive index=1.33

ch0	ch1	ch2	ch3	ch4	ch5	ch6	ch7	ch8	ch9
58	42	0	0	0	0	0	0	0	0
0	9	32	32	25	0	0	0	0	0
0	0	0	0	10	50	40	0	0	0
0	0	0	0	0	0	50	50	0	0
0	0	0	0	0	0	0	66	33	0

Table II
Probability Table for refractive indices=1.33,1.38,1.44,1.48,1.54,1.60

ch0	ch1	ch2	ch3	ch4	ch5	ch6	ch7	ch8	ch9
47	52	0	0	0	0	0	0	0	0
0	9	24	24	22	14	4	0	0	0
0	0	0	0	4	26	47	21	0	0
0	0	0	0	0	0	9	51	39	0
0	0	0	0	0	0	0	32	49	18

Table III
Probability Table for refractive indices
1.33,1.38,1.44,1.48,1.54,1.60
1.33-0.01i,1.38-0.01i,1.44-0.01i,1.48-0.01i,1.54-0.01i,1.60-0.01i

ch0	ch1	ch2	ch3	ch4	ch5	ch6	ch7	ch8	ch9
34	45	3	3	9	3	0	0	0	0
0	7	20	21	18	15	10	4	2	0
0	0	0	0	2	17	32	21	7	14
0	0	0	0	0	0	2	12	14	0
0	0	0	0	0	0	0	19	29	20

From these tables you can see increasing complexity but that it depends on the refractive index as well as size. How then do you derive a probability distribution that can be used in the setting of boundaries?

I would recommend that instead of trying to do an elaborate re-setting of the thresholds, that the thresholds stay as they are and uncertainties in the size thresholds should be estimated based upon the range of expected refractive indices. In figures 1 and 2 below, I have done this in two ways:

Figure one is the average difference between the diameter of particles that should fall in a particular channel, D_{actual} and the diameters that actually fall in that channel from the upper channels. These are based on the probability tables like Tables 1-3. For example, The average difference between Channel 0 diameter and those that actually fall in that channel when the refractive index is 1.33 is 0.42*(3.5-2.5) = 0.42 μm. Channel 2 has a much larger error because the average error is

Diameter error = 0.32*(4.5-3.5) 0.32*(5.5-3.5) 0.25*(6.5-3.5).

Figure 1 shows these average differences as a function of diameter for different mixtures of refractive indices. The overall effect is that the size distribution will always be shifted to smaller sizes. Figure 2 is the same as Figure 1 except expressed as a percentage of the size expected in each channel.

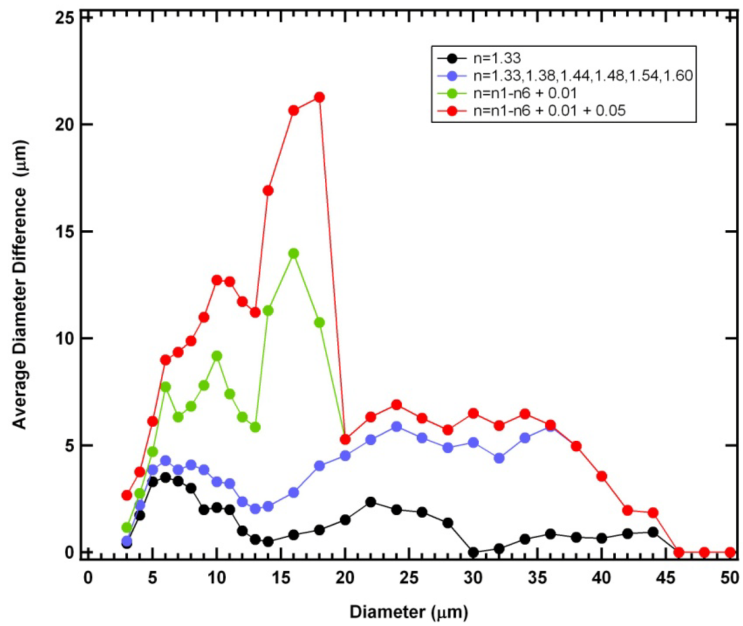

Figure 1

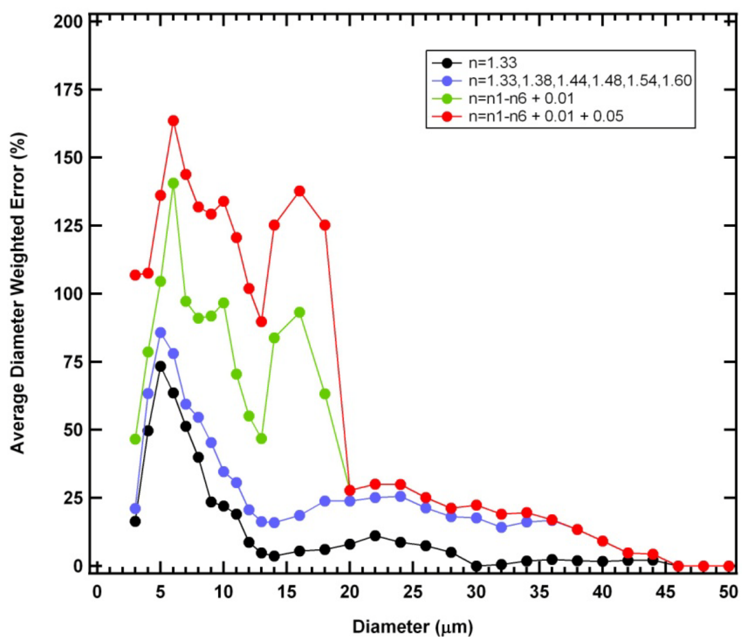

Figure 2

If the software that the authors have developed could produce error tables like these, the users of the data could then establish bounds on the subsequently derived variables like surface area or volume that are needed to evaluate how the aerosol population impacts radiative forcing (surface area) or the life time of the particles (surface area and mass).

Response Functions

I mentioned previously that the authors should consider a more rigorous approach to evaluating the size distributions, such is done with SMPS data. With probability tables like those shown in Tables 1-3, there is the opportunity to invert the data and arrive at a better estimate of the size distribution than by trying to set new size thresholds and bin widths, i.e. given the actual size distribution **A,** the measured distribution, **M**, and the distortion matrix (probability table) **T**, then **A=TM** if the transformation matrix is correct. This is the classic inversion situation that has been solved in various ways.

I would strongly suggest that the authors pursue this approach.

Minor Comments

“Shadow probes” usually referred to as imaging probe or optical array probes (OAP).

Page 103, last sentence. As I am aware of the particle by particle format from the CDP, I am aware of what is being described here but doubt anyone else will. Suggest expanding and explain what is meant here and its relevance.

In section 2.2.3 and 2.2.4, it was never clear to me why scanning or multiple points are needed. If the instrument’s amplifiers are linear, then the relationship between the measured light intensity and subsequent digitize counts should be linear and only a single point would be needed to establish the relationship between counts and cross section. As I point out earlier, multiple points are needed to derive the optimum collection angles.

Section 3 – The mathematics are impressive but without some type of definitive example that shows how these number are derived and applied, you’ll lose most of the readers.