Response to reviewers' comments

We would firstly like to thank both anonymous reviewers and Darrel Baumgardner for their comments which have certainly improved the quality of this manuscript. All their points are listed below with point-by-point responses. We hope that the suggested changes will satisfy any of the reviewers' concerns. In the responses below the reviewer comments are in bold, responses in ordinary text and any suggested new/modified sections of text for the manuscript are in italics. Finally large additional /replacement sections of text and figures are provided at the end.

There are also some general points which we would like to make regarding the reviewers comments and how we have addressed them. There were a number of reviewer comments regarding either too much detail given or requesting more detail in specific aspects of the manuscript. In all cases changes will be made to the manuscript to address these comments. Figure 5 came under significant scrutiny and a replacement figure is included at the end of this document. We believe that this revised figure will ensure the points made are clearer and alleviate many of the concerns the reviewers had with using a DMA.

There were also some questions regarding whether the data quality of the in the field measurements had been improved using this method, specifically with the potential introduction of peaks into the data and comparison with the true distribution. We have performed a significant amount of work since the publication of the discussion manuscript based on the reviewers' comments and personal communications from others who have read the paper. Three items have resulted here.

1) We discovered a hitherto unknown change to the CDP default setup by the manufacturer during a past hardware upgrade. Essentially the bin boundaries of the CDP were not the values we understood them to be. Using the correct bin boundaries in our calibration has significantly improved agreement between the PCASP and CDP.
2) We have performed a first order analysis of the impact of misalignment on the instruments as suggested by Baumgardner. Access to the operational instruments and performing of maintenance tasks since the Fennec campaign has prevented us making measurements of any alignment offsets, however, we have now added what we consider a typical alignment uncertainty into our uncertainty analysis. The additional uncertainty has had limited impact upon the CDP measurements effecting mostly the larger bins, but the uncertainties of the PCASP over its whole range have been increased.
3) Although there is obviously no "true measurement" of the size distribution during the Fennec field campaign we are now able to compare the OPC measurements with that of a cloud imaging probe which images particles with 15 micron resolution. A size distribution is then derived from these images. The size range of this instrument overlaps with the CDP and the comparison is favourable. As this instrument uses a totally different measurement technique this comparison increases confidence in our methods

Based on these changes a revised figure 8 and section 4 have been written and are included at the end of this document.

It is clear from some of the reviewers comments that we have not explained well enough some of the principles upon which the paper is based. Particularly when converting between diameter and cross section space we generate a probability density function (PDF) based on the parameter mean and uncertainty, transform this PDF to the new parameter and use it to define an expectation (best guess) and uncertainty of the new parameter. Baumgardner stated this explicitly in his review and it is clear that a statement to this effect should be included in the manuscript's introduction and abstract.

We would also like to draw attention to the comment of Darrel Baumgardner **...of far greater value (in this reviewer's opinion), is to give those who use the data more tools for estimating uncertainties.** This is very much our view and we have placed traceability at the core of this work. This has enabled us to put meaningful error bars on the in-situ size distribution plots which we feel has been difficult to do rigorously to date and a key output of this manuscript.

Reviewer Comments

Reviewer 1

Change all submicron and super micron to sub-micrometer and super-micrometer respectively.
This will be done

7/27 Obscure sentence
Replace this sentence "For the first... ...on pulse height" with
The CDP provides a histogram of pulse heights with 30 bins every second. In addition it provides the incidence time and pulse height at maximum instrument resolution for the first 256 particles detected per second.This is known as particle-by-particle data.

9 There is no need to go into the details of the DMA technique here. The DMA has been used extensively before to calibrate OPCs, e.g. Covert, D. S., Heintzenberg, J. and Hansson, H.-C.: Electro-optical detection of external mixtures in aerosols. *Aerosol Sci. Technol.* 12, 446-456, 1990; Covert, D. S. and Heintzenberg, J.: Size distributions and chemical properties of aerosol at Ny-Ålesund, Svalbard. *Atmos. Environ.* 27A, 2989-2997, 1993; Okada, K. and Heintzenberg, J.: Size distribution, state of mixture and morphology of urban aerosol particles at given electrical mobilities. *J. Aerosol Sci.* 34, 1539-1553, 2003.
This section will be significantly shortened, although we will retain Eq.[1] and [2] as they are referred to later.

10/25 Where do the numerical values of the weighing function *w* come from?
These are derived from the optical geometry of the probes as given by the instrument specification. Reviewer 2 also commented on these values and Baumgardner suggested that instrument-to-instrument variation should be discussed. For clarity they will be added to a table similar to below
Tabe 1. Nominal Optical Collection Angles for a PCASP and CDP

Instrument	*PCASP*	*CDP*

Direct beam collecting angle(˚)	*35-120**	*4-12**
Reflected beam collecting angle (˚)	*60-145**	*N/A*
Overall weighting function, $w(\vartheta)$	*1 for $35<\vartheta<60$* *2 for $60<\vartheta<120$* *1 for $120<\vartheta<145$* *0 otherwise*	*1 for $4<\vartheta<12$* *0 otherwise*

**Nominal values based on manufacturer specifications. Instrument-to-instrument variation is discussed in sect.4.1.*

Section 4.1 with the uncertainty discussion can be found at the end of this document

11/13 What are the many references meant to substantiate here?
They are the sources of the RI values used to calculate the curves in Fig. 1. The references shall be moved to the figure caption and their purpose stated explicitly.

12/2 How do you justify the choice of a Gaussian function, which is not self-evident to me, in particular at the lower end of the size-sensitivity curve of an OPC where the sensitivity varies considerably with particle diameter?
See the general comments above regarding transformations between diameter and cross section. Here we are dealing with the uncertainty in a particle's diameter and how to propagate that uncertainty when calculating its scattering cross section. Providing the uncertainty in diameter ΔD_p is normally distributed (which is generally the null hypothesis for uncertainty calculations) then a Gaussian is the correct choice. The varying sensitivity you describe is I think what we have described as the variation in cross section with diameter which is taken account of in the $\sigma(D_p)$ terms of Eq. (4) and (5). Basically we are describing here is a more rigorous method of converting from diameter to cross section by taking a probability density function (PDF) of the mode diameter, transforming this into cross section space and calculating a new expectation and standard deviation. This is an alternative to taking $D_p \pm \Delta D_p$, calculating $\sigma(D_p)$, $\sigma(D_p+\Delta D_p)$, $\sigma(D_p-\Delta D_p)$, finding that the three values are not equidistant (or worse that both errors are on the same side of the mean!), worrying that one might be on a spike/trough in the curve and trying to generate some "quick and dirty" value for the cross section and its uncertainty.

17/8 Dealing with the multiply charged fraction of calibration particles downstream of a DMA is not quite as trivial as presented here. Only if an OPC can clearly separate singlets and multiplets (which I'd like to see for the present study, the doublets in Fig. 5 are not connected to any individual singlet) they can be corrected for. This correction only works if the size-sensitivity of the OPC is the same for both, the singlets and the multiplets, which not necessary is the case, in particular at range boundaries.
Reviewer 2 also had some concerns about Fig . 5 and a replacement for this figure can be found at the end of this document. A clear separation can now easily be seen between the singlets and multiplets.
Regarding the size sensitivity I assume the reviewer refers to the fact that as the resolution of the probe coarsens towards larger sizes the peaks can merge together. This is something we have observed and is the reason why we don't use the DMA up to its full 1 µm range in the scanning calibration. Below 0.5 microns we generally have no problems distinguishing peaks with the PCASP.
19/20 This approach had been used several times before (cf. references above)
The Lance et al. paper was chosen because it refers specifically to a CDP, however, it is correct that earlier work be referenced so the earlier of these three papers (covert et al 1990) will be added.
23/4 The "complex" method does not relieve the OPC user from diameter uncertainties in the case of unknown refractive index, particle homogeneity,

shape and orientation.
It does not. However it does provide a framework for propagation of these uncertainties if the user can quantify them. The method has now been extended to allow uncertainties in the scattering curve, whatever their source, to be propagated into the final solution. We have used this method to propagate uncertainties due to optical alignment and will add an extra subsection *3.1 Uncertainties in scattering properties and curves* which can be found at the end of this document and will add additional information to section 3 to communicate the maths involved

Reviewer 2

General Points

1) The work that this manuscript is based on is for sure not to underestimate. Nevertheless it seems that it is incomplete. The authors describe a calibration method and apply this on a measured data set. The result they compare with the same data set treated according to the specifications of the instrument producer. Why do the authors provide no absolute comparison with a reference. Then it would be much clearer which technique is better. In this context it is not clear why the authors used a DMA for generating particles but they did not use a SMPS for checking the DMA generated aerosol size distribution. At least a CPC in parallel to the calibrated PCASP could provide something that can be compared with what is recorded by the PCASP – this would improve the described calibration procedure by additionally calibrating concerning OPC-measured particle number.
We are not entirely sure to what the reviewer means by an absolute comparison. The data shown in Fig. 8 are from an aircraft sortie during a field campaign in the Central Sahara. Unfortunately this means there is no absolute reference for comparison. There were other OPCs on board which are not discussed here as they had not been through the same calibration process and there was an imaging probe which has a range overlapping with the CDP measurements. This is discussed further in the reviewer's specific comments. The only measurement of goodness we can utilise is consistency between instruments. This has now been found to be very good based on the comments found at the beginning of this response and the replacement for Fig. 8 at the end of this document. During calibration we have sometimes used a CPC with the DMA for checking concentrations measured by the PCASP and, depending upon the OPC in question, this is a useful thing to do and we have included this in an additional section 1.3 found at the end of this document . The problem of OPC counting efficiency is a difficult one in its own right and we specifically named the paper "Particle sizing calibration" because we do not elaborate on counting efficiency. Some difficulties of using a CPC to investigate counting efficiency are:

1) It can only be used alongside a DMA so that small particle contamination of the sample below the detection threshold of the OPC can be screened out.
2) It does not tell you about the inlet efficiencies and turbulent losses at aircraft speeds.
3) It increases the flow rate in the DMA, broadening the DMA peaks. This is discussed further in the reviewer's general point 3)
4) It may not be possible in the field when such equipment is not available.

We are not sure how to interpret the reviewers comment regarding using an SMPS to check the DMA as an SMPS is essentially a DMA with a scanning voltage source and its output connected to a CPC. Unfortunately cost also limits equipment availability and transport of radioactive materials legislation precludes loaning such a piece of instrumentation so an SMPS is simply not available for our use.

2) I'm wondering about the use of the expression "particle size distribution" in connection

with Figures 3 and 5 and in according caption and text referring to the figures. What is shown here is anything but a particle size distribution. What is shown here is a pulse height in unspecified units versus a frequency of appearance (by the way, without any unit). So, I suggest that established termini are used more carefully. It would help, just to change into another expression.

Yes this terminology has been used loosely and will be replaced with *particle response distribution* or simply *particle distribution* where appropriate. The plot shows no units because it has been normalised and is therefore unitless – again as you indicate this precludes it being referred to as a particle size distribution. The replacement for figure 5 has had the y axis labelled as *relative units* for clarity.

3) The authors mention turbulences in an inlet, particle losses in tubes, the bandwidth of the DMA selecting certain particle-electromobility-diameter (which is not the true particle diameter). But it is not very clear how these uncertainties are considered in the calibration and particle sizing.

These topics will be addressed further in the reviewer's specific comments, but are quickly addressed here. Turbulence affects concentration measurements during aircraft use and tubing losses affect concentration measurements in the lab, however, for narrow distributions these losses have little effect on the shape of the measured distribution and hence on the sizing calibration. The statements about short tubing are made simply to indicate we are following good practice and will be removed as they are not entirely relevant.

The DMA peak width defines the slope of the sigmoids in Fig. 6 and hence has some impact on the uncertainty in the best fit parameters, but this impact is weak so is not discussed. A comment on this relationship will be added. Most importantly, however, is to ensure the peak widths are narrow enough that the gap between singly and multiply charged peaks can be resolved. This has already been discussed in the comments of reviewer 1.

For spherical particles the electromobility diameter is a good approximation to the true diameter, this is validated during calibration of the DMA with PSL spheres and is the reason why we suggest use of PSL spheres and DEHS oil, both of which produce spherical particles.

We will clump some of these extra uncertainties together in a subsection entitled *Further measurement uncertainties* provided at the end of this document

4) Some sentences are pretty long (e.g. page 109, 12-15; page 118,16-19 – more cases can be found throughout the text), additionally with quite complex constructions. The manuscript would improve if these constructions were "streamlined".

Agreed. We will streamline the sentence structure as much as we can.

Specifics: Page 99, line 25: of which size, concretely, are the "largest particles"?

The factor of three applies for particles of 120 micrometres diameter and above. The largest particles seen were 120 micrometres although the CDP had potential to see particles up to 170 micrometres. This particle size will be defined in the manuscript

Page 100, line 6: I guess "though" should be "through"

This will be changed

Page 100, lines 9-10: why once writing 0.06m and later "one-hundred micrometers"?

This will be changed so both use numerical representation

Page 100, line 12: What is meant with "Shadow OPC", please provide an example and Reference(s), if available.

We will now use the term imaging probe, rather than shadow OPC as recommended by Baumgardner. As we will now make use of some data for an imaging probe a fuller description will be included. We will remove the text "Shadow OPCs provide size distributions up to mm sizes, but are not discussed further here." From Section 1.1 and a fuller description is added to a revised section 4 to be found at the end of this document.

Page 100, line 19: In which sense is "particle is homogeneous" meant?

Composed of the same material throughout – please see next comment

Page 100, line 21: In which sense is "homogeneous water" meant?

Perhaps this is where the confusion arose, because water is clearly homogeneous. A hyphen will be added to give *homogeneous water-particle* making it clear that it is the water-particle that is homogeneous, not the water itself

Page 101, lines 1-3: Please provide Instruments names and Reference(s), if available.

The sentence "some OPCs collate... ...finest resolution allowed by the electronics" will be replaced with

Some OPCs collate particle events into discrete time and/or pulse height bins(including the Grimm OPC (Heim et al. 2008)) while others provide the time and pulse height for every particle at the finest resolution allowed by the electronics(for example the SID2 (Cotton et al. (2010)).

Page 101, lines 22-23: A word seems to be missing in this sentence.

This sentence will be reworded as it is not very clear.

Page101, line 27: please provide concrete values for the sample and sheath flow of the PCASP in the setup that is used here

These will be added here for clarity and wording changed slightly when they are referred to later in the paper

Page 102, lines 12-13: knowing these digit values is pretty useless for a reader

They will be removed

Page 102, line 19-21: the description of the inlet is not very clear. It seems that the authors mean a diffusor-type inlet. Perhaps following references help to specify this specific inlet:

"Wilson et al., Stratospheric sulfate aerosol in and near the northern hemisphere polar vortex: The morphology of the sulfate layer, multimodal size distribution, and the effect of denitrification, J. Geophys. Res., 97, 7997–8013, 1992" or

"Hermann, et al., Sampling Characteristics of an Aircraft-Borne Aerosol Inlet System, J. Atmos. Ocean. Tech., 18, 7–19, 2001."

It is a diffuser type inlet but not quite like either of the two described in these references due to differences in how the subsampler is aligned and how the excess flow exits. The wording will be changed to include the word diffuser and a description is added to the new section entitled *Further measurement uncertainties.*

Page 102, line 22: how does this needle valve impact particle losses?

The term needle inlet is used by the manufacturer and is perhaps confusing. There is no needle valve involved and the term probably originates from the fact that the inlet is long and thin like a hypodermic needle. All references to this term will be removed to avoid confusion.

Page 102, line 18-28: This will cause turbulences and thus additional particle losses

Yes it does, as mentioned later in the reviewer comments this is discussed later in this paragraph. We will group this information together in a section called *Further measurement uncertainties* to make things tidier.

Page 102., lines 28-29: In which sense is the subsample maintained? For a reader that does not know the work of Belyaev and Levin it would help if the "range covered" is provided explicitly.

The word maintained is perhaps misleading as there is no active control. This sentence will be replaced with: *This ensures that as aircraft speed increases and pump efficiency decreases with increasing altitude, the ratio of subsampling speed to the inlet speed remain within the range 0.18-6.0 covered by Belyaev and Levin (1974).*

Page 103, lines 2-9: Now a statement about the turbulence comes up - and a bunch of additional information is following. Seems not to be essential for understanding the

calibration described later. Apart from this any conclusive statement at the end of this section is missing. Bottom line: these details about the aircraft operation, increased flow, turbulences, fluid-dynamics, etc. leaves more questions coming up than it answers.

As you say, this is not essential in terms of calibrating the PCASP's sizing ability and brings up more questions than answers. Unfortunately these are unresolved questions and this was intended as a flag to the user that there are some unresolved characterisation issues with these instruments. For the sake of neatness and clarity these items will be put together in a separate subsection as mentioned earlier.

Page 103, lines 20: "known as the Sizer and the Qualifier" is there a reference defining these expressions?

These are terms used by the manufacturer and also in the Lance 2010 paper. They are described in the proceeding paragraph. The Lance paper will be referenced here.

Page 104, line 2-3: This statement is seductive for asking the authors: Why not? This would even further upgrade the manuscript.

It certainly would improve the manuscript, but is probably enough work in itself to warrant a separate paper. We are therefore not proposing to add it to this manuscript. One of the co-authors is currently planning to undertake this work in the future.

Page 106, line 16-22: I suggest to put all of this in a list placed underneath the equation.

This will be done

Page 106, lines25-27: For the sake of clearness I suggest to put this in a table.

This will be done, see proposed Table 1 above

Page 108, line 13: Which type of DMA? Are there any references?

The DMA is described in more detail later. The phrase "*the DMA*" will be replaced with "*a DMA*" so there is no implication that the reader should already be familiar with it.

Page 109,line 1: how is the particle number controlled? Was any reference particle counter used?

The control is a manual process with the operator pressing a valve to release compressed air. Faster airflow generates higher concentrations. The number concentration is monitored on the CDP itself. We will add *manually regulated* and replace "to the CDP" with "*as measured by the CDP*"

Page 109, line 5: At which concrete particle concentration would coincidence be expected to cause problems for the CDP?

Coincidence bias is obviously a continuously varying property of concentration, so it is difficult to put an absolute figure on it. Lance et al (2010) found that concentration measurements were unaffected by coincidence up to concentrations of ~100 cm^{-3}. Correcting for aircraft speed this works out to be ~500 s^{-1}. Note however that the mode of a distribution should be more robust to coincidence than the absolute concentration. We will add reference to the Lance paper and state this figure.

Page 109, lines 6-7: So how are described problems avoided in this work?

They aren't. As described later we do not use small particles in the calibration because of these problems. This unfortunately increases uncertainty in the smaller range of the CDP. For future work we are looking into using PSL spheres instead, perhaps replace our calibration beads more frequently, add regulated flow control to our set up and/or filter out data where concentrations are high. This will be explicitly stated here.

Page 109, lines 11-13: how long is the tube length, concretely, if talking about "directly connected", or "a section of flexible tubing".

Page 109, line 16: please specify "as short as possible"

In both cases 20-30 cm. The impact of this is small due to the particle sizes and flow rates used but it just highlights good practice. As mentioned previously we will remove reference to tube lengths.

Page 111, lines 4-5: Neither the expression in the Fig 3 caption is correct nor the text referring to Fig 3. This figure shows pulse height vs frequency and not "mode particle scattering cross section". The Figure is not a particle size distribution. One is confused about the number in the graphs: what is the mode diameter

The quote "mode particle scattering cross section" does not refer to Fig. 3, but to the processes used to derive Fig 4. To ensure this is clear we will replace the sentence "To generate a... ... measured by the OPC." With

To generate a calibration equation for the OPCs the mode particle scattering cross section and associated uncertainty for each of the calibration particles is derived using Eq. (4) and (5) and these are plotted against the equivalent modes in the particle distributions from Fig. 3

As mentioned earlier particle size distribution will be replaced with particle response distribution or particle distribution. The "mode diameter" is the mode diameter of the calibration particles used to create each of the particle response distributions in Fig 3. This is now stated explicitly in the above rewording.

Page 113, lines 8-12: The particles-inertia based impactor technique is not the best way to "physically remove" multiply charged particles from a calibration aerosol.

Both TSI and Grimm supply impactors with their DMAs for the purpose of removing multiply charged peaks so this is certainly one option, however care must be taken that the cut off diameter is appropriate for the size being used. We only use data where the singlets and multiplets can be resolved so this is simply a hint for the reader if they cannot resolve multiplets.

Page 114, lines 12-14: This sentence makes obvious that particle number was measured somehow. And one would like to know with which instrument that is not shown in the calibration setup and how/where these measurements were used or why not used.

The concentrations here are from the PCASP itself. This will be made explicit.

Page 119, lines 14-15: The argumentation of not well defined lower boundaries of the first bins should be discussed in more detail to make plausible that there is a rational to simply discard measurement data.

The PCASP has the lower boundary of its first bin defined by the pulse width rather than the pulse height in order to effectively screen out electrical noise. It is difficult if not impossible to associate this pulse width with a diameter. We had thought the same was true for the CDP but it seems we were mistaken and the lower limit for the CDP is defined, just not in the same part of the data logging software as the other bin boundaries. Bin 1 of the CDP is now included in the analysis. The lower boundary of the CDP was not set to the manufacturer's recommended setting, but was much lower. No adverse effects (e.g. electrical noise) were observed, however the manufacturer's estimate of the diameter equivalent of this bin bottom is clearly not valid. Data from this bin is, however, used in the calibrated data set.

The sentence "It should be noted that the first bins... ...not have well defined lower boundaries" will be replaced with

It should be noted that the lower edge of the first bin of the PCASP is defined in terms of a pulse width rather than a pulse height to reduce the impact of electrical noise. This means that the range of bin 1 is not easily defined and the data from this bin is discarded.

Page 121, lines 8-10: From this section one gets the impression that more "corrections" were applied to the data set which the new calibration technique is applied on. In contrast, it seems that one took less care of the data based on the manufacturer specifications. If so, the data sets cannot be compared that easy. If not, the procedure should be described more carefully and detailed.

Three corrections were applied to the calibrated data. The calibration itself, the RI correction and the gain stage edge correction. The third of these had limited discussion in the rest of the manuscript so may seem to have come from nowhere. We will add an additional section between sections 2 and 3 entitled *PCASP gain stage boundaries* to describe this correction more fully.

Page 121, lines 15-16: Nevertheless the discontinuity is still present. Is one of aims of this new calibration procedure really to reduce this discontinuity? Where is the proof that the reduced discontinuity is closer to the true size distribution? On the other hand features like a mode between 1.5 and 4 m are amplified. In the panel "c" one could misinterpret this as a mode in the particle size distribution – this risk would be minimized

in the size distribution shown in panel “a”.

The source of the discontinuity in the calibrated RI corrected data has been located. As discussed in the introductory paragraphs the incorrect pulse height definitions of the bin boundaries were used. This error has been rectified and the agreement is now much better as can be see in the new Fig 8 at the end of this document. We believe that two well calibrated instruments should agree within their uncertainty (at least 3-sigma uncertainty) and I think that it is clear that this is not the case for the manufacturer spec (although the manufacturer does not provide uncertainties). However, we don’t believe it is fair to expect they would agree as the specifications assume different refractive indices to that of the aerosol being measured. The curves are presented here simply to show the net effect of the work in this paper upon the size distributions and to show that if materials are measured that are not those assumed in the manufacturer’s specification then good results can still be achieved. The mode that you refer to may be a residual of the scattering function used which has a number of inputs. We have now assessed the impact of misalignment upon the PCASP and this has increased the error bars on this mode. It may be that it is an artefact of the incorrect refractive index or it may be that the use of Mie theory is not a perfect assumption. As detailed in the manuscript assessment of the scattering phase function of dust is beyond the scope of this paper. Of course the possibility exists that this could be a real mode.

The distribution from the CIP particle imaging probe has now been added which agrees very well with the CDP data set and increases confidence that we are getting a good representation of the true distribution. Of course it is impossible to prove without doubt that data from a field campaign is the truth, however, this work provides a traceability and transparency of assumptions that is not the case with data using the manufacturer’s bin boundaries.

Page 122, lines 1-3: A reader could ask why this work was not completed by the data from the shadow OPC.

At the time of submission the final dataset was not available. The data is now available and is included in the replacement Fig. 8 at the end of this document

Figures: Figure 1: Suggestion: Change the colors of the graphs such that one color is not used for two different substances.

Where a colour has been used twice it is to indicate a range of scattering properties for the same substance as described in the caption

Figure 3: The ordinate axis has no unit. It is not clear where the numbers (in units of m) in the graphics come from. Finally the question arises if these are single measurement runs or if the experiments were repeated several times. In later case, a statistic (error bars) would be nice.

The ordinate axis has no units due to the normalisation as described in the caption. The label will be replaced to state *Number / bin width (relative units)* These are from a single calibration run.

Figure 8: The label “d” is missing in according graph. The comparison would improve if “c” and “d” were shown together with “a” and “b” in one plot, respectively.

The replacement figure 8 at the end of this document combines the plots as described

Figure 5: The lines between the measurement points might cause misinterpretation. In fact the measured data point should be shown without any line in between. Furthermore the question arises where the 0.30m in the graphic is connected to. If this value fits to the maximum that is locate exactly underneath the 0.28m peak the question comes up how this can be?

A replacement Fig. 5 is now provided at the end of this document wihich is plotted as a bar chart histogram. It is split into multiple rows, labelled appropriately so no confusion can occur. The reviewer is correct in his association of labels to peaks. Two sizes can give peaks in the same channel due to the coarse resolution of the instrument at in this range. It is now clear in the bar charts that the adjacent bins to the peak receive some particles and there is some variation in these bins as would be expected as the peak moves to larger diameters.

Figure 6: The ordinate label “F” should be highlighted. Error bars are missing here.

Error bars are of course important, but may make this already busy figure unreadable. A statement about uncertainty will be added to the caption

Reviewer 3 Darrel Baumgardner

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.

The other reviewers have made similar comments. The DMA description will be shortened and Mie theory section will be shortened. We, however, feel that because Mie theory is relied upon strongly in the rest of this paper some introduction is necessary to keep the paper self-contained. In particular we feel Eq (3) is important as it essentially sets up the problem we are dealing with, and Eq (4) and(5) are important as they begin the definition of our uncertainty propagation. We will change the list of parameters of Eq (3) to a list for better clarity. The definition of w(θ) will be put in a table and based on your comments later uncertainties will be added to a later section.

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.

Since submission we have also been examining issues relating to the probe collecting angles varying from nominal. As I'm sure you are aware in addition to the limiting values of θ being incorrect by a constant value, the sample may be displaced laterally as discussed in Lance et al. (2010) meaning that the limiting values of θ become functions of ϕ. We have found that the impact of a lateral variation is to fill in some of the troughs in the Mie-Lorenz curve for deviations up to 2 degrees. We will be working to perform the measurements you describe, however, due to limits upon access to these instruments which are used operationally, the timescales for this work are not suitable for publication in this manuscript. Instead we have examined the impact such deviations have on the final data set and have included these in our uncertainty estimates and will present this methodology in a new section 3.1 to be found at the end of this manuscript.

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.

Regarding the polarisation, the instruments are unaffected when they collect uniformly for all ϕ which is the case for the PCASP and CDP, at least with the nominal alignments. It is the case, however that for some other instruments which collect over finite ranges of ϕ that polarisation could have an impact and this will be highlighted in the text and with changes to Eq (3).

The broadening due to Gaussian mode lasers this will be explicitly stated also.

Measurement uncertainties related to refractive index

I have not repeated the section of text, tables and figures here, please refer directly to the reviewer's comments for these. However, in summary, the reviewer presents an idea whereby instead of redefining the equivalent diameters of each bin the equivalent diameters remain constant and an error is defined (referred to by the reviewer in his equation simply as "Diameter error" and referred to in the rest of this section as $\text{Error}_{\text{Baum}}$) which indicates the mean difference between the diameter of particles which fall in the bin and the mean diameter from the manufacturer's specification.

It seems that the reviewer uses the word error here in its literal sense, i.e. to mean a know offset rather than an uncertainty and this is then used as an input in his later Matrix Inversion suggestion.

This error could be accounted for using

correct_bin_mean=manufacturers_bin_mean+$\text{Error}_{\text{Baum}}$

In fact if the resolution is increased in the derivation of $\text{Error}_{\text{Baum}}$ such that we calculate it over small intervals of diameter or cross section ranges then correct_bin_mean as defined above becomes equivalent to $\overline{D}_{b_{perfect}}$ in Eq (11) of the manuscript. In this case Fig. 7 shows equivalent information to one row of the reviewer's table, but at higher resolution.

It therefore appears that we have begun from the same starting point as the reviewer but rather than using this to define an instrument response matrix as in the reviewer's next comments we have increased the resolution to generate an integral and derived procedures defining bin centres and means and for error propagation.

Given that the reviewer has essentially described the principles upon which our refractive index corrections are based we are happy that we seem to be using sensible methodology and don't intend to make any changes to the manuscript based on these comments.

Response Function

Another method proposed by the reviewer is that of a matrix inversion problem. Again please see the review for full details. Where we have used the information in Fig. 7, and added uncertainty analysis to generate information about the instrument bins in diameter, the reviewer suggests instead that we define an instrument transformation matrix or kernel **T** such that the real size distribution **R** generates a measurement **M** via

RT=M

The problem then condenses to finding $\mathbf{T}^{-1}$.

We considered this a very interesting proposal and could think immediately of one potential advantage of this method. When using $\overline{D}_b$ to generate a size distribution there is an implicit assumption that each of the sub-bins used to generate $\overline{D}_b$ contain equal concentrations. This causes a broadening of the distribution as the actual mean of the bin will lie closer to the distribution peak than the value calculated. In the matrix inversion method the assumption is that the concentration across each bin of **R** is equal. Because the bins in **R** are continuous so cover a narrower range of diameters, this is a better assumption than used in the derivation of $\overline{D}_b$ so should cause less broadening.

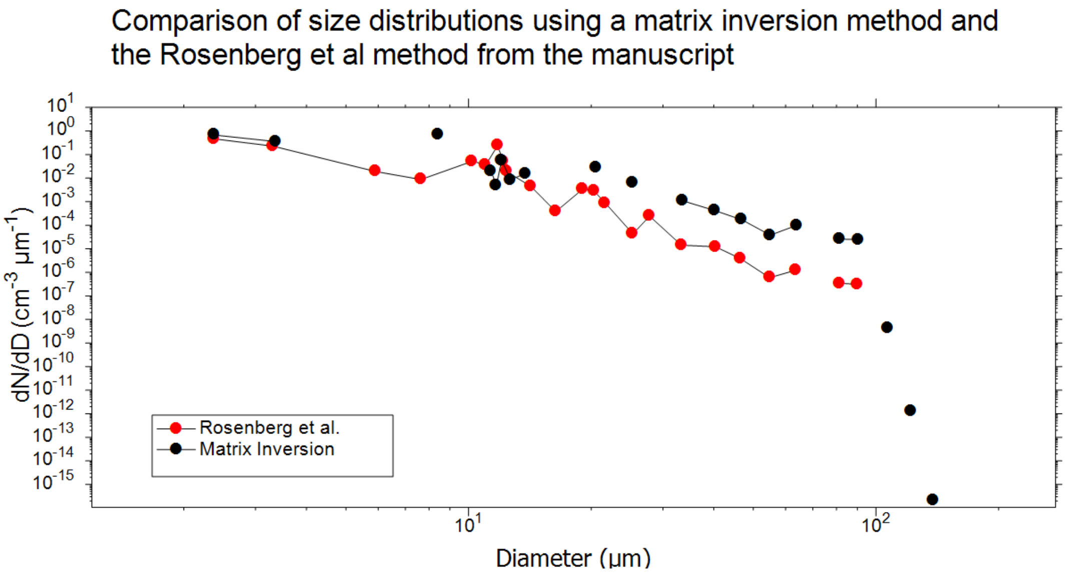
We have therefore tested this method using the same example as in the manuscript for the CDP. Unfortunately we came across a number of difficulties listed below.

1) **Selection of appropriate bins for R**
   The boundaries of the bins of **R** are an input to this algorithm. When similar matrix inversion methods are used with e.g. DMA measurements, steps are picked in the voltage that ensure overlapping of peaks in the transfer functions. This gives obvious choices in terms of the bins to use in **R**. With a Mie-Lorenz curve there is no opportunity to set up the instrument response to provide obvious choices in this manner. We have found that simply using the Manufacturer's specification results in a kernel **T** with some identical rows and zero columns meaning it has no inverse. This is the case for the Fennec data, but in general there is no reason that any manufacturer's bin boundaries should be suitable, especially if the refractive index used is different to that expected by the Manufacturer or if there has been a drift in instrument sensitivity. A method which selects bins for **R** by maximising the diagonal elements of **T** would give a matrix which had a better chance of being invertible. Such a method would define steps in diameter for **R** which are approximately equivalent to the steps in the cross section boundaries of **M**. These steps have already been defined by $W_{b\ perfect}$ from the manuscript's Eq. (11) and (12). Indeed using steps in the boundaries of **R** defined by $W_{b\ perfect}$ gives a matrix which is invertible. We therefore find that The matrix inversion method is not independent of, but reliant upon, the methods described in the manuscript.
2) **Propagation of uncertainty in concentration**
   As we have uncertainties in our knowledge of the instrument kernel we must propagate these through into the real size distribution **R**. We envisaged this being possible as follows:
   a. Vary the bin boundaries in terms of cross section, and assign a weight to each variant based on the uncertainty in the boundary and the distance of the variant from the mean.
   b. Generate a new version of **T** and its inverse for each variant
   c. Generate **R** for each kernel variant and use the spread and weightings of the results to define an uncertainty and a mean R.

   Unfortunately it may be the case that not all the variants of **T** are invertible giving a situation where it is not possible to effectively propagate uncertainties. We found that this was the case and in the example given below, basing the bin boundaries on W_b rather than $W_{b\ perfect}$ gave a **T** which was not invertible.

3) **Definition of uncertainty in diameter**
   As described in 1) above the bins of **R** are inputs into the algorithm. Hence no uncertainties can be specified and no indication of the instrument's sizing uncertainty can be given.
4) **Numerical resilience**
   We found that when we used $\mathbf{T}^{-1}$ to generate a size distribution as shown below, negative concentrations existed in some bins (indicated by gaps on the log scaled size distribution). Presumably because the transformation should conserve total number this caused a large increase in concentration in some remaining positive bins. There are also some spurious points added when concentration drops to zero. We believe these problems are likely to originate from the fact that the inversion is not resilient to differences between the calculated value of **T** and the "real" instrument kernel. If the ratio of particles in some bins is not consistent with **T** due to calibration uncertainties, counting errors or uncertainties in the

scattering function/refractive index of the measured particles then the method can create negative concentrations.


Comparison of size distributions using a matrix inversion method and the Rosenberg et al method from the manuscript

Unfortunately although this method initially seemed to be promising we find that its lack of resilience and inability to effectively propagate uncertainty make it unsuitable for use with OPCs. We will add a statement to the manuscript to this effect.

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

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.
This sentence was commented upon by another reviewer and will be reformatted as detailed above. To expand briefly another 2 sentences will be added
This means that particle grouping can be examined or that the particles can be rebinned after logging has taken place. In this way much more information is available and the data is more flexible.

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.
Even for linear electronics offsets can exist so at the absolute minimum 2 data points would be needed. Obviously each data point has an associated uncertainty so the error in the resulting straight line is reduced by adding additional data points. In section 2.2.4 no assumption is made regarding the instrument electronics. This method can be used with nonlinear electronics or in situations where the pulse heights cannot be directly measured.

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.

Fig. 7 shows an example of how Eq (11) and (12) work. $\overline{D}_{b_{perfect}}$ and $W_{b\ perfect}$ will be added to this plot to indicate it's direct relevance. In addition a Fig. 7b will be constructed to show a similarly represent the uncertainty propagation in Eq (13)-(16). Eq. (18) is simply a bivariate normal distribution so will be condensed into a similar form as Eq(17).

Additional/replacement sections and figures

New section 1.3 Further measurement uncertainties

Particle sizing is only one aspect of the function of an OPC. The other is particle concentration measurement. Although this work does not deal directly with this aspect of calibration it is useful to consider some of the problems which may be encountered with the data presented here. For a closed path instrument with an inlet such as the PCASP, the representativeness of the concentration measurement is often known as the sampling efficiency and is 1 for perfect sampling, less than 1 for undersampling and more than 1 for oversampling. For an open path instrument such as the CDP the concentration measurement relies upon defining a sample area, also known as the depth of field. The sample area defines a cross sectional area of the laser beam through which passing particles will be counted. Multiplying the sample area by the airspeed and time interval provides a sample volume allowing a concentration to be measured. Lance et al. (2010) showed that the CDP sample area can be measured using a droplet gun on a micropositioning.

Despite the fact that PCASPs or other instruments with identical inlet systems have been flown on aircraft for decades there seems to be a dearth of measurements of the PCASP sampling efficiency at aircraft speeds. In the laboratory an OPC can be compared with another standard instrument such as a condensation particle counter. This has been performed in the past with the FAAM PCASP and agreement is within 20% for all sizes. Application of such an efficiency is non-trivial to transfer to aircraft measurements. This is because of the high speed airflow from which we are sampling. The PCASP samples initially through a diffuser inlet which is aspirated via ram pressure from the aircraft motion. This has a conical shape with a cross section initially of 70 mm^2 (diameter 9.4 mm) increasing to 10 times this value. The mean sample velocity is reduced correspondingly and a small subsample is drawn through another inlet with cross section 0.05 mm^2 (diameter 0.25 mm). The remaining excess sample exits through a vent tube on the side of the PCASP. Belyaev and Levin (1974) provided empirical corrections for sampling efficiencies when sampling from a moving airstream in which the ratio of airstream velocity to inlet velocity is in the range 0.18-6.0. The PCASP subsampling rate has been set to 3.0 cm s^{-1} on the ground in order that the flow ratio remains within these limits where possible (note the aircraft speed increases and the subsampling speed decreases with increasing altitude). The data presented here assume an inlet efficiency of 1 for the diffuser and then assume sampling efficiencies based on the mean flow speed at the subsampling inlet and the Belyaev and Levin (1974) relations. It should be noted, however, that flow inside the diffuser is expected to be turbulent as the Reynolds number at the tip may be as high as 60,000 during flight. A 3-dimensional incompressible fluid dynamics model with direct numerical simulation of turbulence has confirmed that flow separation occurs in the conical inlet leading to turbulent eddies at the subsampler. Further investigation in terms of compressible fluid modelling, inlet comparison and wind tunnel testing will be required to assess the impact of this turbulence on the sampled size distribution. Likely effects include turbulent losses in the diffuser and errors in inlet efficiency of the subsampling inlet

New section 3 Gain stage boundaries of the PCASP.

As described earlier the PCASP uses three separate gain stages to maximise its range. If a pulse saturates the first gain stage it is passed to the second. If it saturates this gain stage it is passed to the third and if it saturates the third gain stage it is registered as oversized. Where the first gain stage overlaps the second gain stage it reduces the width of the first bin of the second gain stage. This is because some particles that would be measured by the second gain stage do not saturated the first gain stage so are instead counted in the top bin of the first gain stage. A similar process occurs where the second and third gain stage overlaps. After performing the calibration detailed in Sect. 2.2.3 independently for each gain stage the limiting boundaries of the gain stages must be compared. Where an overlap occurs the bottom of one gain stage must be set to the top of the previous gain stage.

Unfortunately, despite this correction, size distributions from the PCASP tend to show concentrations which are too high in the top bin of each gain stage and too low in the first bin of the second and third gain stage. It is suggested here that particles are not correctly registering as saturated so are getting stuck in the top bin of a gain stage. To investigate this problem the PCASP was reprogrammed to zoom in on the overlap region between the mid and low gain stages (medium and large particles). The particle distributions as a function of scattering cross section for the two gain stages are shown in Fig A2. A number of unexpected features are evident here

1) The concentration in the second gain stage remains zero for some distance beyond the overlap point.
2) The last bin in the first gain stage has enhanced concentrations.
3) The enhancement in the last bin of the first gain stage is of a similar order of magnitude (approximately 50%) to the depletion in the second gain stage.
4) The concentration in the last bin of the low gain stage is significantly enhanced.

In addition the concentration of oversized particles is only 0.028 cm^{-3} which is much lower than expected given the concentration in the top bin of 2.47 cm^{-3}, and the bin before this of 0.30 cm^{-3}. This plot seems consistent with our hypothesis that particles are getting stuck at the top of a gain stage and are not effectively moving to the next gain stage or being classified as oversized. At the very least some undocumented process is affecting the distribution at the gain stage boundaries. Unfortunately the mechanism causing this problem is not known, however, an effective workaround is to merge the bins either side of each gain stage boundary and discard the final bin of the PCASP.

New Section 4.1 Uncertainties in scattering properties and curves – to be added to what is now section 3 but will become section 4

The uncertainty propagation presented thus far has assumed that the scattering curve which is generated using Eq. (3) is a perfect representation of the response of an OPC to a particle of a particular size. In reality the weighting function w(θ,ϕ) will have an uncertainty associated with it as will the refractive index of the particles being measured. For particles which deviate from perfect spheres the assumption of Mie-Lorenz scattering or use of a different scattering function may also introduce uncertainty. As will be detailed in Sect. 5, the impact of refractive index and particle shape has not been studied here, however the variation in probe geometry and its input into the instrument uncertainty has been examined. Because for both these probes sampling is symmetric about the laser axis we consider two possible deviations from nominal.

1) A simple change in the limits presented in Table 1. This could represent a deviation from nominal of an aperture or a movement of the sample volume along the axis of the laser. Note that for the PCASP only the 35˚ and 145˚ limits are varied as these are most sensitive to

the position of the laser/sample intersection point. For the CDP the total angular range is maintained at 8˚ by altering both limits by the same amount. This is referred to as an along axis deviation.

2) A change in the centre point of the optics away from 0˚. This could represent a movement of the sensitive volume perpendicular to the laser axis, e.g. due to imperfect laser alignment. Again only the 35˚ and 145˚ limits of the PCASP are considered. These estimateswere made using a four point integration around the laser axis. The four points were perpendicular to the deviation (where change from nominal is approximated as zero) and parallel to the deviation (where change from nominal is maximised). We refer to this as a lateral deviation.

Measurements of the position of the laser beam of the PCASP during alignment have shown that a maximum lateral deviation of 1 mm can be expected. A similar uncertainty is expected for the along axis deviation. Both these misalignments give changes in the 35˚ and 145˚ collecting angle limits of approximately 10˚. For the CDP Lance et al (2010) found that a lateral deviation of 1.4 mm gave the best fit to calibration data, this equates to a deviation of ~2˚. Consideration of an along axis deviation was not presented in that work. Baumgardner (2012) reported that the manufacturers of the CDP have begun testing the responses of these instruments to small particles in order to estimate the collecting angle of the instrument. They have found maximum deviation of the lower collection angle limit of 0.7˚. A higher variation was found for the upper collection angle limit, but this has less impact upon the instrument sensitivity. As the majority of these numbers are maximum offsets of a relatively small population of measurements they have been assumed here to be 2-sigma estimate. Therefore the 1-sigma uncertainty in collecting angles of a typical PCASP and CDP used in this work have been assumed to be 5˚ and 0.4˚ (rounding to 1 significant figure) respectively for along axis deviations and 5˚ and 1˚ respectively for lateral deviations.
For the PCASP we found almost no variation in response to desert dust for lateral deviation of 1-sigma. 1-sigma along axis deviation did, however, lead to a significant change in response. For the CDP lateral deviation did induce some changes in response, but these were smaller than for along axis deviations. Mie-Lorenz curves for these cases are presented in Fig A2. Because in both cases the along axis uncertainties dominate we shall consider only these here. It should be noted, however, that these conclusions are valid only for the refractive index in question. For the CDP the variation in signal due to misalignment was found to be much smaller for glass bead calibration particles and water (not presented here) than for dust.

Revised Section 4 (now section 5)

In June 2011 the FAAM aircraft was deployed to the Sahara to make dynamics, radiation and dust measurements. The PCASP and CDP were employed to make measurements of particle concentrations and size distributions of desert dust and cloud particles and a part of this dataset is presented here. Prior to this campaign the PCASP and CDP were both calibrated using the discrete method described in Sect. 2 and the CDP was calibrated using the same method before each flight. Unfortunately a step change in the gain of the high gain stage of the PCASP is thought to have occurred between calibration and the beginning of the project and hence the first 6 bins of the PCASP have not been included here. It should also be noted that the first bins of the CDP and PCASP are routinely discarded as they do not have well defined lower boundaries.

As Fig. 7 shows, the actual ranges from a PCASP bin can vary significantly from the values provided by the manufacturer. In the calibration performed before Fennec the bin centres were found to be systematically higher than those reported by the manufacturer by an average of 13 % and a maximum of 33 %. Monitoring the calibration results over approximately 1 year has shown that after routine maintenance, such as cleaning and aligning the optics, the calibration may change by up to

20 %. This result is consistent with the 35˚ and 145˚ limits of the PCASP collection optics varying by up to 10˚ as discussed in Sect. 4.1 The drift over time is typically much less than this and calibrations performed before and after projects which have lasted a month or more show less than 5 % drift.

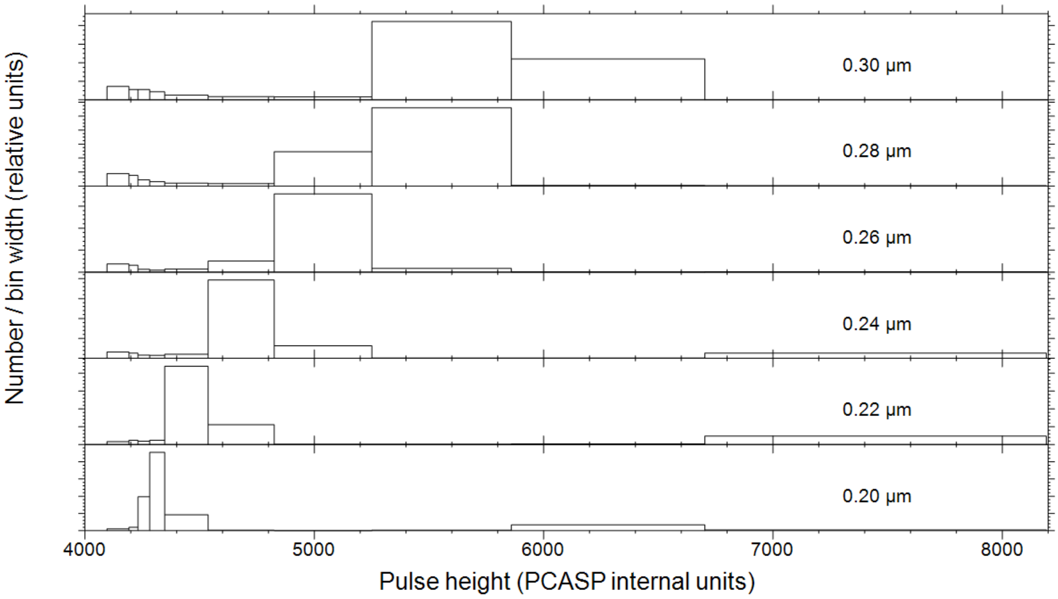
During Fennec the CDP was calibrated before every flight except one and these have been examined to check the stability of the instrument over this time period. It was found that for all bins of the CDP the drift over the project was less that either the 2-sigma uncertainty of the calibration or 9 %. The difference compared to the manufacture's nominal boundaries was found to be in the range 1.2 to 3.7 µm diameter.

Size distributions from one time period during the Fennec project are shown here. This case consists of 150 seconds of data beginning at 10:10:30 UT and collected at 800 m above the surface (1080 m GPS altitude). This was a measurement period with particularly high dust loadings. There is some uncertainty in the refractive index and shape of the dust measured and here it has been assumed that the dust particles are spheres with a refractive index of 1.53+0.003i which lies in the range measured by Wagner et al. (2011). Laboratory measurements have shown that Mie-Lorenz calculations can have some success in modelling the scattering properties of non-spherical particles. In the forward scattering angles as measured by the CDP laboratory measurements of bulk desert dust samples, including Saharan dust, agreed with Mie-Lorenz calculations within 20 % when surface area equivalent diameters were used (Volten et al 2001, Kahnert et al. 2007). The scattering cross sections of ~0.2 µm salt particles as measured by a PCASP were modelled by Mie-Lorenz theory to within experimental uncertainties when mean crystal length equivalent diameter was used (Lui et al. 1992). It is beyond the scope of this paper to evaluate the many scattering theories which have been applied to nonspherical particles and hence based on the successes above Mie-Lorenz theory has been used.

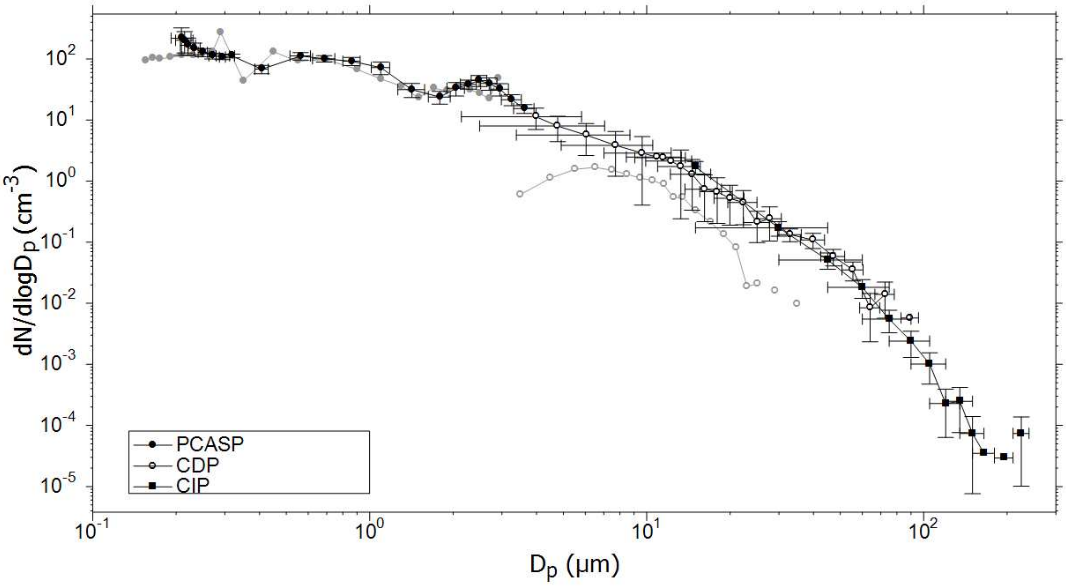
The number and volume distributions as a function of particle diameter for the described time period are shown in Fig. 8. Distributions are compared using the manufacturer's specifications and calibrated, refractive index corrected bin boundaries. The distributions using the manufacturer's specification are discontinuous at the boundary between the two instruments around 4 µm and the PCASP data shows a zigzag in the distribution at 0.3 µm (the boundary between the mid and low gain stages) and a peak in number concentration in the last channel as described in Sect. 3. A similar zigzag is usually seen at the high to mid gain boundary at around 0.14 µm. The gain stage boundary corrections described in Sect. 3 have been applied to the calibrated data set

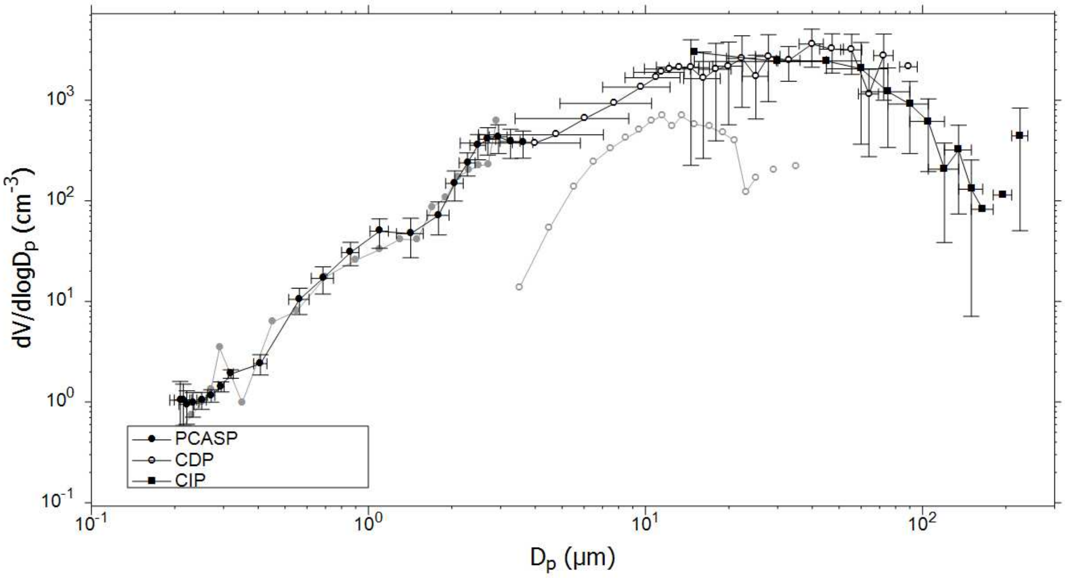
The calibrated data can be seen to extend to much larger diameters than that processed using the manufacturer's specification. This is mostly due to the impact of the different refractive index of the measured dust and the PSL spheres and water droplets referenced by the manufacturer. The two instruments are in excellent agreement where they meet and any discontinuity is much less than the 1-sigma error bars plotted. The calibration procedure combined with the CDP's particle-by-particle feature described in Sect. 1.2 could allow the data to be rebinned by redefining the bin boundaries as seen fit in terms of internal instrument units. Here the manufacturer recommended bin boundaries have been maintained except for the first 5 bins which extend below the size of the CDP's normal 1st bin. It could be considered that the bumps seen in the PCASP have been accentuated by the calibration and refractive index correction presented here, perhaps especially so for the mode between 1 and 2 µm. It could be the case that this is a real mode or there is the potential that this is an artefact caused by imperfect knowledge of the particle scattering properties. The error bars at this point are a significant fraction of the mode height so the statistical significance of this peak is not clear. Although amplification of such a peak may be considered a negative of this method the addition of error bars to this plot which are traceable and transparent is seen as a significant benefit.

In addition to the OPC data, Fig. 8 also shows data from the Cloud Imaging Probe (CIP), which was part of the Cloud, Aerosol and Precipitation Spectrometer (Baumgardner et al. 2001) operated during Fennec. The CIP is an optical array probe (also known as an imaging probe) as initially described by Knollenberg (1970). The instrument directs a laser at a linear array of photodetectors and when a particle travels through the laser, perpendicular to the array, its shadow is imaged line-by-line. Utilisation of data from this instrument provides a comparison with a completely different particle sizing technique. Because the CIP, CDP, and PCASP all agree very well and the uncertainties in the measurements appear to be representative of the observed variations we have high confidence that our calibration and refractive index correction methods work well and that the uncertainty propagation is effective.
It is of note that the Fig. 8 does not symmetrically bound the mode, despite data being available for particles as large as 200 µm. It is clear that the volume distribution of desert dust can have contributions from particles larger than have previously been measured on an airborne platform.



Replacement Figure 5. Particle response distributions showing the concentration of particles in each bin measured during a scanning calibration of a PCASP using a DMA. Each bin is normalised by dividing by its width. The labels indicate the mode diameter of the singly charged peak of the DMA output, D*. The resolution here is not as good as in Fig. 3 as the bin boundaries used here are those for normal use; no zooming is applied. Only the mid gain stage is shown. Doubly charged peaks can be seen in the D* = 0.20, 0.22 and 0.24 µm plots, but are smeared by the course PCASP resolution at this size.





Replacement Figure 8. Size distribution of desert dust aerosol measured by the PCASP, CDP and CIP during a straight and level run at 800 m above the surface. Plots show the distributions derived using the manufacturer's specification based on the refractive index of PSL spheres and water for the PCASP and CDP respectively (grey) and the distributions derived from calibrated refractive index corrected data (black). The number, N, and volume, V, are shown as a function of particle diameter D$_p$. Error bars which extend to negative numbers on the log scale have been omitted for clarity.

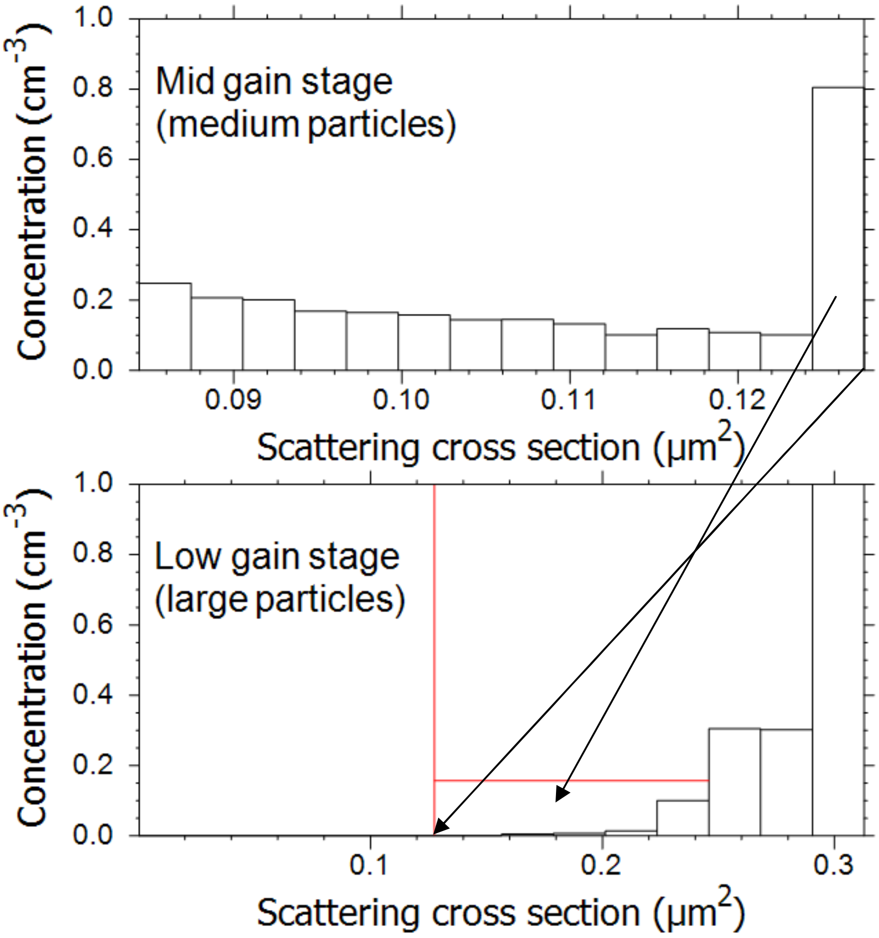


Figure A1. Particle distribution at a PCASP gain stage boundary. Plots show details at a PCASP gain stage boundary created by reprogramming a PCASP to zoom in on this area of interest. The red vertical bar shows the maximum extent of the overlap between the two gain stages, below which we expect to see no particles. The horizontal red bar shows the concentration which would be measured if the excess in the top channel of the mid gain stage were redistributed above the overlap point of the low gain stage. Note that the top bin of the low gain stage goes off scale to a concentration of 2.47 cm^{-3}.

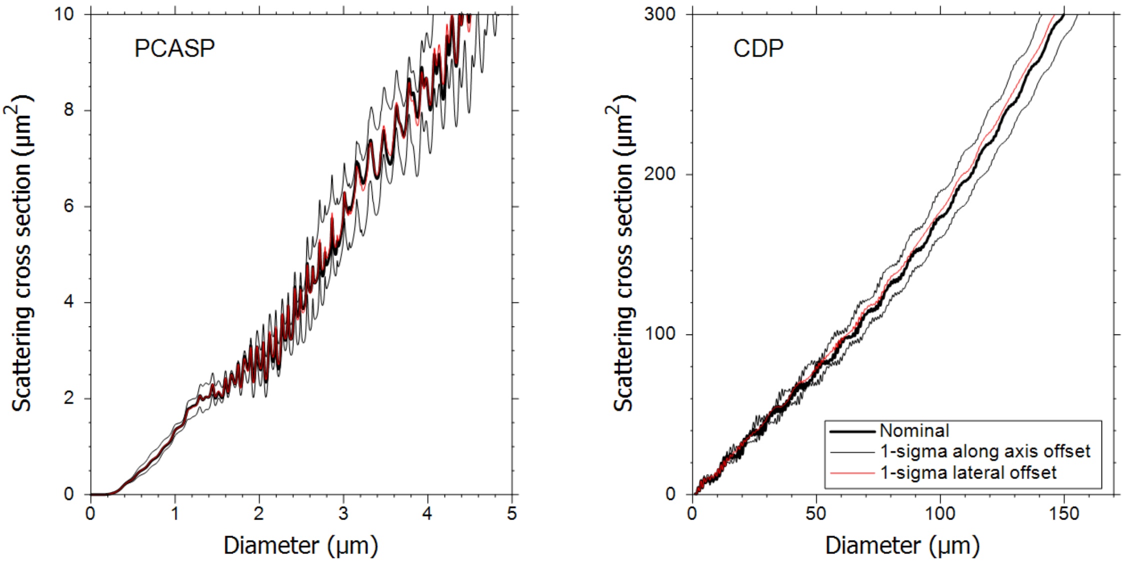


Figure A2. Mie-Lorenz curves for the PCASP and CPD showing the impact of misalignment of the optics for desert dust. The thick black line shows the scattering cross section measured by the instruments using the nominal manufacturer's specification. The thin red and black lines show the impact of moving the sample/laser intersect point or sample volume along the laser axis or laterally in a direction perpendicular to the laser axis. The 1-sigma offsets are estimates of the variation from nominal for a typical instrument and are based observed offsets of FAAM's PCASP after realignments and measurements of a number of CDPs by the manufacturer (Baumgardner et al. 2012).