Reply to the reviewers comments and changes to the manuscript

„An optical particle size spectrometer for aircraft-borne measurements in IAGOS-CARIBIC, Atmos. Meas. Tech. Discuss., 8, 11597-11642, doi:10.5194/amtd-8-11597-2015, 2015

We thank the two reviewers for their comments, which helped to improve the manuscript. Minor comments, like missing words or spelling errors, were all correct according to the reviewer's suggestions. Major comments and questions (in italic) are addressed below:

Comments by reviewer # 1 (Darrel Baumgardner)

1. *Are there no publications that describe this instrument and compare it with other techniques like SMPS, UHSAS, etc.?*

   To our knowledge the RION KS-93 has not been used for atmospheric research before, and there are no respective publications about KS-93 measurements or comparisons with other aerosol particle measurement systems. After the CARIBIC OPSS was built up, we did a quick comparison with an UHSAS for ambient laboratory air. The distributions where similar, however, the absolute numbers in the CARIBIC OPSS where higher, probably due to a flow control problem in the CARIBIC OPSS at that time. As we were pressed for time, unfortunately we did not do a comparison with one of our reference SMPS systems. But this could be done in the future, for instance during the CARIBC container modification at the end of this year. It should be noted that the CARIBIC OPSS is now part of the CARIBIC measurement container, thus we usually have only access to the instrument for calibration once or twice per year, for a few days only.

   Since end of 2014, an SP2 instrument is operated inside the CARIBIC container in parallel to the OPSS. A comparison of the particle size distribution measured with both instruments (although limited by the SP2 in particle size range) is in progress.

2. *I think that either Fig. 1 or 2 should have a scale beside it that shows the relative size. I am also not convinced that both figures are needed.*

   We agree with the reviewer that Fig. 1 and 2 are somewhat redundant and removed Fig. 1. Furthermore, we added a scale to the former Fig. 2, now Fig. 1, as requested.

3. *This sentence* (in Sec. 2.2 on page 11602 starting in line 20) *is incomplete or needs to be rewritten for better clarity.*

   The respective sentence was rephrased:

"Besides the modified KS-93, the CARIBIC OPSS houses electronic components (EMI filter, DC/DC converters) and a flow control system (mass flow controllers, compressor, as well as temperature and pressure sensors)."

4. *Not sure what this means. From 1 to 3? Hard to distinguish in the figure.*
   In Fig. 2, the ports of the valve were renamed, the font size for the numbers was increased, and the respective sentences clarified "In the stand-by mode (green arrows in Fig. 2), about 3 l min-1 of filtered air originating from the diaphragm compressor leave the two-way selector valve via port 1." and "In measuring mode (blue arrows in Fig. 2) the two-way selector valve is switched and the particle-free air from the compressor is now directed through port 2. At the same time the external vacuum pump is switched on."

5. *If it is constant volumetric flow, why does the pulse duration vary so much?*
   Our text seemed to be a little bit misleading. As you can see in Fig 4 (b) and (d) the full width half maximum for both particles (200 nm and 900 nm) is about 30 µs, hence the pulse duration is not different. The numbers given in the text refer to the original, not amplified signals. For the original signals the pulse duration difference comes from the three different amplifiers. We clarified this by changing the respective sentence (where actually the 90 µs were wrong, it should be 150 µs, see Fig. 4 c) and adding an additional sentence:

   "As the original pulse of a single particle has a duration of about 60 to 150 µs (see Fig. 4), each pulse is thus resolved with 20 to 50 data points. The full width half maximum of the amplified particle signals is about 30 µs for all particle diameters."

6. *Why such high frequency sampling for housekeeping?*
   The chosen real-time data acquisition system is a little bit oversized, it has more performance than actually needed. Hence, the housekeeping data are sampled with such a high frequency just because we can easily do it, but it would not be necessary for the CARIBIC OPSS operation.

7. *What would be the typical file size? Can the information on every pulse be recorded, i.e. is the transmission rate of the data sufficiently fast so that even at high number concentrations every piece of data is transmitted with no lost particles?*
   It seems that the paragraph at the end of Sec. 2.3 was formulated not well enough. The OPSS particle data are only stored in the OPSS, the communication with the master is only for unit control reasons and follows a simple protocol with a few bytes of ASCII text per command. To clarify this, we added an additional sentence at the end of Sec. 2.3:

"The OPSS communication with the master is restricted to simple control commands and answers, whereas the particle signal data are solely stored in the CARIBIC OPSS."

8. *I don't understand this sentence.* (about the trigger level)

   We hope to clarify the respective sentence by changing to:

   "The trigger level of Ch1 was set to 0.35 V for the amplified signal as for measurements of particle-free air and using a lower trigger level of 0.30 V small particle pulse signals were recorded. These artificial particle signals are caused by electronic noise."

9. *Should you add here that post processing shape analysis of the pulses might allow additional filtering of these spurious particles?*

   This information was added:

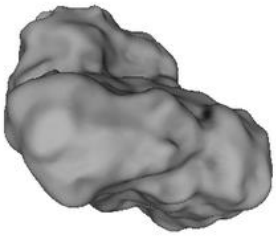
   "For such an application more sophisticated criteria, like post-processing shape analysis of the pulses, might allow appropriate filtering."

10. *T-matrix? Aspect ratio?*

    The program for calculating the OPSS response functions for non-spherical particles was developed in PhD thesis at TROPOS and the detailed description of the program will we part of a different paper. Nevertheless, we added additional information about this program in the text and two papers in references:

    "Therefore the UT/LMS response function was calculated again for slightly irregular-shaped particles with moderate surface structure (Pfeifer, 2014). The calculations are based on discrete dipole approximation (DDA, e.g., Yurkin and Hoekstra, 2007) and were conducted with the ADDA program (Absorption by Discrete Dipole Approximation) from Yurkin and Hoekstra, 2011. The respective response function is displayed in Fig. 8b."

    To let the reviewer know, how the particle we used to calculate the non-spherical response function, looks like (aspect ratio), we include a picture here in the reply. However, we do not want to display this picture in the paper, because the reader might think that "all particles" in the UT/LMS look like this particle. But this is not the case, non-sphericity could be one reason, but there can be others, as already stated in the text.



Particle shape used to derive the non-spherical response function in the manuscript.

11. *I don't understand how such a large peak can be smoothed by the non-spherical response function. Where are those particles being sized?*

    The difference between the spherical and non-spherical response function is particularly strong in the particle size range 700-1000 nm, see Fig. 8b. In this size range the amplified particle signal is interpreted as a particle up to 200 nm smaller when using the non-spherical response function compared to the particle diameter obtained using the spherical response function. This large decrease in particle diameter leads to the very large decrease in particle volume for the second peak in Fig. 9, as the volume scales with the particle diameter cubed. Obviously the OPSS derived particle volume distribution is very sensitive to the particle diameter and the associated uncertainty in the volume distribution is large.

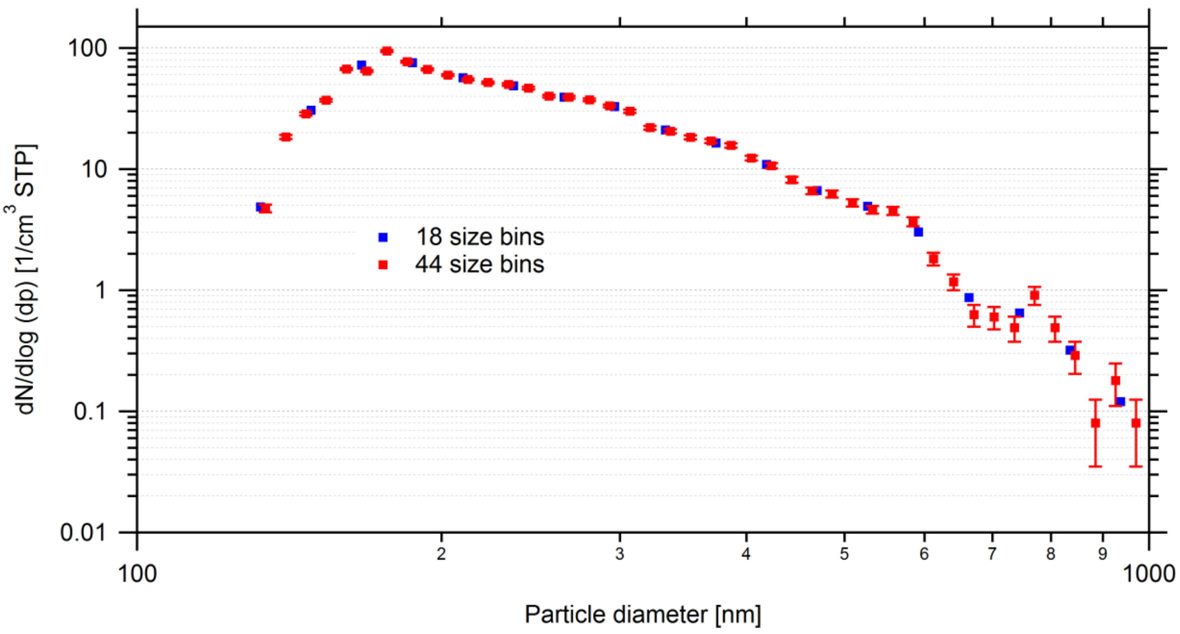
12. *I have trouble with the justification here since it seems what you are trying to do if find a "nice" response function that doesn't have a nasty plateau. Since you have particle by particle data, you can actually test your assumption that these are slightly aspherical. Create a size distribution with many more intervals than 10 using your response function and you should see if indeed the response function is smooth. If it is, than in the interval between 500 and 700 um the curve should be smoothly decreasing, assuming a typical accumulation mode spectra. If not, there will be an unsightly inflection caused by the fact that the response function is not monotonic.*

    In the PhD thesis, this work is partly based on, you can see that initially, we used a very simple, “two straight lines plus interpolation” response function, which did do quite a good job. However, then we stumble over the small, but partly high peaks in the particle volume distribution, which we do not believe to be real. This was the reason for modelling the response function for spherical as well as non-spherical particles.

    As we state in the respective paragraph, we do not claim that the measured particles were non-spherical, there can be other reasons, which can cause a similar response function to the one we finally use, e.g., a changing refractive index with increasing particle diameter. To emphasize this, we now added two additional sentences in the conclusions:

    “The non-spherical particle response function was finally chosen for data evaluation, as it leads to smoother particle volume distributions. The use of a non-spherical particle response function is no proof that the majority of the UT/LMS particles are non-spherical. There can be other reasons which lead to similar response functions, for instance unknown properties of the optics or a particle size dependent refractive index.”

Finally, we performed the exercise suggested by the reviewer. In the figure below you see a particle size distribution measured during flight LH339, the flight we discussed the volume distributions in Fig. 9. The two curves are based on the same particle data, however we used two different size bins numbers. The figure shows that increasing the number of sizes bins does not change the shape of the size distribution in the range of the first Mie resonance, predicted between about 580 and 710 nm (see Fig. 8b). There seems to emerge a feature slightly above that region, at 770 nm, however, considering the statistical uncertainties (here displayed as error bars only for the size distribution with the larger number of bins in red) the curves are equal for most size bins. We interpret this as that there might be a small plateau in the real response function, but this is above predicted the size range of the first Mie resonance, and it is so small that, for the finally chosen number of size bins (10) it is not resolved and thus not important.



Two particle size distributions based on the same particle data but using two different OPSS size bin numbers (18 and 44 respectively). Error bars (only display for the red data points) give the statistical uncertainty (Poisson). For the blue squares these uncertainties are smaller because of the larger bin width.

13. *CPCs are also susceptible to coincidence and a correction factor typically applied. Is this being taken into account here?*

No, the CPC data were originally not corrected for particle coincidence, because the overall coincidence effect for our measurements was considered to be (and is) small. To be exact, the CPC data are now corrected for particle coincidence and Fig. 11 was updated. The resulting new coincidence numbers were implemented in the text accordingly:

“The coincidence effect is below **4%**“ (old value 3%) „for an ambient particle number concentration of 1500 cm^{-3}. Considering that, based on CARIBIC measurements, the ambient accumulation mode particle number concentration in the UT/LMS rarely exceeds 100 cm^{-3} (coincidence below **0.25**%)“ (old value 0.2%) „ the raw data were not corrected for coincidence in the post-flight data processing.”

The CPC coincidence correction does not change the original statement, for our UT/LMS measurements particle coincidence in the CARIBIC OPSS does not play a role, in particular when considering other uncertainties, like the inlet transmission or the unknown refractive index.

14. *Maybe the concentration of saturated pulses would be a useful parameter to tabulate as well? What is the cut-point of the aerosol inlet?*

    Yes, we used the information of particles with saturated pulses already in another paper (Rauthe-Schöch et al., 2012) to extend our measurements into a size range not resolved by the OPSS. This bears of course large uncertainties, because the exact shape of the size distribution above 1 µm and the inlet transmission in this size range are not well known. The upper threshold diameter (50% transmission efficiency) of the CARIBIC inlet can be estimated according to data obtained for similar inlets to be around 3-5 µm. This is well above the 1.05 µm upper limit of our OPSS data.

15. *Can you be more specific about where and when the OPSS measurements were made? These are averages from 2010-2013? Over what latitudes?*

    For the comparison with the balloon-borne measurements data from two periods were used. This is described in more detail now, see below. The latitude and altitude band was already provided in the text, all data south of 65°N which fall into the 150-700 ppbv ozone altitude range. We detailed this information a little bit by changing to:

    “For the comparison two periods of little volcanic influence (according to Andersson et al., 2015) were chosen, from June 2010 to May 2011 and from May 2012 to June 2013. Data from these periods are considered representative for the background aerosol. The balloon and CARIBIC particle size distribution data sets in these two periods were restricted to measurements between 30°N and 65°N and to the LMS (ozone values between 150-700 ppbv), as the LMS aerosol shows much less variation with sampling location than the UT aerosol (see Fig. 13)."

16. *It also seems like a very large time gap from the Wyoming measurements and the OPSS. How many volcanic eruptions, large forest fires etc. occurred since the Wyoming measurements?*

    Due to the relative low measurements frequency of the balloon measurements there are of course large time lags (weeks) between the balloon-borne measurements and an actual CARIBIC flight. However, we accounted for that by using only data from the LMS, where the aerosol is more homogeneous compared to the UT, and we did a statistical comparison and not compare an individual flight to an individual balloon launch.

    The influence of major volcanic eruptions on the data was excluded by choosing the two periods given above. Minor volcanic eruptions (VEI 4 or smaller) and biomass burning events (e.g. pyro-convection) rarely reach the lower stratosphere. For biomass burning plumes there is an estimate that only 17 events per year world-wide reach altitudes above 8 km (Guan et al., ACP, 2010). Hence it is relatively unlikely that one of the nine balloon flights was influenced by a strong biomass burning event. The OPSS data stem from much more flights and are collected over a much larger area, hence in principle they might be influenced by one or two strong biomass burning events. However, for the representation of the OPSS data we used the median, which is insensitive to such rare events.

17. *How does the refractive index impact the number concentration, other than how it alters the lower and upper size thresholds? I think I understand what you are indicating, i.e. that the concentration in each size channel depends on the definition of the size thresholds that then impacts the normalization. I think that it is worth making this more clear as it is a not so obvious connection.*

    Part of the particle number concentration uncertainty in an OPSS size bin is caused by the refractive index. For a fixed set of size bins (border diameters), different refractive indices and thus different response functions can assign a certain particle signal into different size bins. Thus for one refractive index size bin number two might contain 100 particles, but for a different refractive index only 90 particles. This kind of uncertainty was meant.

    We tried to clarify this statement by expanding the respective sentence in the manuscript to:

“The CARIBIC OPSS uncertainty with respect to particle number concentrations is caused by the uncertainties of the OPSS flow rate (14%, MFC uncertainties) and the OPSS counting efficiency (5% for diameters larger than 200 nm, measurement uncertainty). An additional uncertainty 15% for the individual size bin particle number concentration is caused by the particle refractive index, as different response functions can attribute a given particle signal into different size bins. The above uncertainty was calculated using fixed OPSS size bins, real CARIBIC flight data, and three different response functions. Finally the inlet sampling efficiency for accumulation mode particles contributes to the OPSS number concentration uncertainty with about 10%. This number was estimated from the standard deviation of the wind tunnel experiment particle transmission data of the inlet.”

18. *Root sum square?*

Yes, we mainly used Gaussian error propagation, which is equivalent to root sum square. But this was already indicated in the text, one sentence before the “error propagation”:

“To these randomly distributed uncertainties (Gaussian error propagation) the error caused by the spurious particle signals (below 1%) must be added, whereas coincidence again can be neglected. This error propagation yields a total instrumental uncertainty with respect to the particle number concentration of 19% for the total accumulation mode particle number and 25% for the individual size bins.”

19. *Aren't you forgetting shape?*

Yes, we forgot the shape, although we showed in our manuscript how much shape can matter. We included a new paragraph about the influence of the particle shape on the particle size uncertainty in the discussion, but we cannot provide solid numbers for this uncertainty:

“The particle size uncertainty caused by the unknown particle shape is hard to estimate. Ideally, one would take different original OPSS particle signals, use dozens of different OPSS response functions, each derived for a different particle shape, to transfer the signals into particle sizes, and finally calculate for each original particle signal a particle size probability function. This huge effort could not be carried out in this study. The discussion about the OPSS response function in Sec. 3.4 and the volume distribution in Fig. 9 already indicates, however, that the particle size uncertainty caused by the unknown particle shape is not small for particles in the size range of the laser wavelength or above.”

20. *Are they really monodisperse?* (particles for the OPSS sizing capability determination)

The particles used for the calibration are only quasi-monodisperse, as their distribution width is determined by the respective NIST standard (for latex particles) or by the chosen DMA flow ratio setting (for the ammonium sulfate particles). The uncertainty in the OPSS sizing is therefore partly caused by the calibration particle size distribution width, and the estimate of 6% for the size uncertainty represents an upper limit. We changed the respective statement to:

"The uncertainty of the size calibration can be estimated, using the width of the Gaussian signal distribution when measuring a quasi-monodisperse aerosol. According to laboratory calibrations this uncertainty is 6% for the CARIBIC OPSS (Weigelt, 2015). As the particles used for the calibration are only quasi-monodisperse, their size distribution width causes already some of the spread in the OPSS data, and thus this number is an upper limit."

21. ... *as well as do additional filtering by interarrival time, pulse width and maybe even pulse area in the future.*

We included this additional information and changed the respective sentences in the conclusions to:

"The core instrument, a commercial RION KS-93 particle sensor, was modified to record individual particle pulse signal curves. Due to this high-resolution sampling, the OPSS particle size bin width and the OPSS size distribution time resolution can be adapted at any time after measurement. Moreover, additional filtering by inter-arrival time, pulse width, and maybe even pulse area in the future can be carried out."

22. *I still have a problem with that since it maybe isn't physically reasonable.*

We clarified our statement, as described in the answer to question number 12 in this reply.

Comments by reviewer # 2

23. I am not sure what is novel about this work. The last sentence of page 11600 suggests that this instrument has already been used for particle size distributions, thus I am under the impression that the technique is not new. Is the data processing software new? If I am misunderstanding the text and the instrument is new, what new and cutting edge capability does it provide that makes in more significantly important than previous optical particle sizers? This point was not clear to this reviewer and should be made very clear in a revision. Is the novelty of this paper that just an ordinary OPC was used to collect data in a hard-to-access part of the atmosphere? If so, then the issue remains about what is new about the instrument because this information is needed to qualify this manuscript important for this particular journal.

We apologize that our wording on page 11600 was misleading. The RION KS-93 has, to all of our knowledge and including a literature search in the internet, not been used for atmospheric research before. Up to now, we are the only group using this particle counter for atmospheric aerosol measurements. The three papers mentioned on page 11600 are using the CARIBIC OPSS data, i.e. our instrument, hence they provide scientific results obtained with the instrument described here. But there was no complete description of the CARIBIC OPSS in these papers, and this is done in the manuscript here. To clarify this, we changed the respective sentences on page 11600 to:

"Particle size distributions measured by this OPSS were already used in previous studies (Rauthe-Schöch et al., 2012; Martinsson et al., 2014; Andersson et al., 2015), but here the instrument is described in detail for the first time (Sect. 2). The new OPSS was thoroughly characterized in the laboratory with respect to ..."

The novelty of the CARIBIC OPSS and the manuscript stems from a) the OPSS optics (a synthetic quartz optical cell), which is insensitive to all pressures which might be encountered in the real atmosphere, b) the relative small size and weight of the original OPSS, c) the relative low particle detection limit of 130 nm, which can only be reached by a few other OPSS instruments, d) the possibility to analyze the complete particle pulse signal curve, even years after measurement, and e) the thoroughly characterization of the instrument. Or in the words of the first reviewer:

"*This is not just another OPC in the fuselage of an aircraft. There are many of these on research aircraft that have already been described in the literature. The research team* *has done a very careful analysis of the commercial instrument that they adapted for this application and one of the unique features is the storing of the complete pulse shape for post processing. This offers the possibility of further filtering for artifacts as well as possibly increasing the sizing accuracy."*

Therefore we feel that our manuscript is well appropriate for AMT.

24. *Page 11599, Line 12: I am not sure how this can be expected based on the first part of the sentence "and thus allows the post-flight choice of the time resolution and the size distribution bin width."*.

The CARIBIC OPSS records individual particle signals together with a time stamp. This allows in the postflight analysis to use the particle pulse height, area or even shape. Furthermore, the size bins are not given by the instrument manufacturer, but can be chosen by the operator depending on the application (e.g. using a larger n umber of particle size bins for high particle number concentrations).

We tried to clarify this by changing the respective sentence in the abstract to:

"The instrument records individual particle pulse signal curves in the particle size range 130-1110 nm diameter (for a particle refractive index of 1.47-i0.006) together with a time stamp and thus allows the post-flight choice of the time resolution and the size distribution bin width."

Likewise, the respective sentence in the conclusions was changed:

"The core instrument, a commercial RION KS-93 particle sensor, was modified to record individual particle pulse signal curves together with a time stamp."

25. *Introduction: The authors need to motivate the importance of doing measurements in the "UT/LMS" better. Why are they focusing on this region and not other parts of the atmosphere? What are the major issues right now in the UT/LMS that the community should be concerned about and how can the data generated from this instrument help address those issues? Please add text about this issue.*

As our manuscript describes an instrument, which can be applied also on a mountain station or an aircraft flying in the free troposphere (between 2-8 km at mid-latitudes), we feel that a broad discussion focusing on the UT/LMS aerosol is not appropriate for the introduction. Nevertheless, we added some motivation by adding the following sentences and references to the introduction:

"…for instance in the upper troposphere and lowermost stratosphere (UT/LMS). Aerosol particles in this region provide surface area for heterogeneous chemistry (Søvde et al.,

2007) and influence the formation of ice clouds (Krämer et al., 2009) and thereby indirectly impact the Earth´s radiation budget. The discovery of the Asian Tropopause Aerosol Layer (ATAL) by Vernier et al. (2011) and the much stronger than previously thought radiative impact of volcanic aerosols in the UT/LMS (Andersson et al., 2015) are just two examples for the importance, but also for the limited knowledge of the UT/LMS aerosol."