

Reply to referee 1 for the first part of the LOAC paper (AMT-2015-212)

We want to thank the referee for their nice and detailed analysis of our paper. Please note that the referee's comments are in italic and that our replies are in normal style.

General comments:

The LOAC project is clearly a very ambitious project in view of its versatility, the large range of platforms able to host it, its objective to provide information on aerosol size and type as well, and mainly, over the extremely large targeted particle size range, spanning not less than almost three orders of magnitude. The main question is obviously to determine if this instrument is able to achieve these ambitions. If the extended description of the measurement principle and intercomparisons with other instruments seem convincing for some aspects, it also shows in a recurrent way limitations of the technique for some others, making the performances claimed by the =authors somewhat questionable. Very satisfactory values of uncertainties are provided for some limited aspects of the measurement processing chain (e.g. uncertainty on the counting in well-controlled experimental conditions and for very particular cases of particle size distributions, uncertainties on the laser intensity and on the pumped air flow, standard deviation between a set of successive measurements). But an estimation of the global resulting uncertainty for measurements in real conditions is never given, and the analysis tends to show that many aspects could lead to questionable estimates of the particle concentration: effect of the inlet, which could be poorly controlled concerning the sampling of large particles, bias in particle counting when particles cross simultaneously the laser beam (cf figure 3), insensitivity of the calibration curve to particle size for very thin particles, multiple values of particle diameter corresponding to a given value of the measured "flux" (in mV) around the 3-10 μm particle size range (cf. Figure 4), saturation effects at high concentrations of large particles ($> 10 \mu\text{m}$; cf. Figure 6), etc. Altitude dependence (possibly through temperature/pressure) of the uncertainty is never really tackled. Consequently, LOAC, which seems to give very useful information for the detection of aerosol types, could miss the ambition to provide quantitative estimates of the particle size distribution over a quite large part of the claimed 0.2-100 particle size range. The authors should present their study in a much more critical way to give better and more reliable information over the exact performances which can be expected from LOAC. This is an indispensable condition to bring confidence in the quantitative results which have been provided and will be provided in the future by LOAC.

We have significantly improved the paper, following most of the referee comments. We have also added a new cross-comparison session involving different aerosols counters, in a sea spray chamber using a well-controlled air flow system.

Specific comments:

Abstract

L. 4-5 p.9995: To be precise, in [Deshler et al., 2003, cited in the paper], the authors mention the value of 150 nm as smallest particle size measured by optical particle counters (OPC) at Laramie. However, these authors also measure condensation nuclei (radius $\leq 0.01 \mu\text{m}$) by growing particle with sizes down to 0.01 μm in the sample stream.

We understand the reviewer's comment. But, strictly speaking, a Condensation Nuclei Counter (CNC) is not an aerosol sizer because it does not provide the size distribution between 0.01 μm and $\sim 0.150 \mu\text{m}$. We have changed the sentence to: "Among the numerous instruments available, optical aerosol particles counters (OPC) provide the size distribution in diameter range from about one hundred of nm to a few tens of μm ."

L. 5-6 p.9995: "sensitive to nature of the particles": Do the authors refer to the value of the refractive index? This could be mention for the sake of clarity.

We change "nature" to "refractive index"

L. 23-24 p.9995: The authors should remove the sentence "All these tests indicate that no bias is present in the LOAC measurements and in the corresponding data processing." See the specific discussions further in the text.

We understand the referee's concerns; we have removed then sentence in the abstract. We now provide new cross-comparisons, we have improved the discussion (see below).

1. Introduction

L. 9-10 p.9996: The authors should also mention their role in ozone photochemistry, e.g. through the formation of PSC.

We have changed the sentence to: "In the middle atmosphere, aerosols play a significant role in ozone stratospheric chemistry, including the formation of polar stratospheric clouds, through heterogeneous reactions with nitrogen and halogen species".

L. 15 p.9996: The residence time is not always short ! As the authors will know, aerosols from the Pinatubo eruption in 1990 left a significant signature in the stratosphere at least for 5 or 6 years! Hence, aerosols may be used as a tracer in some circumstances.

The Pinatubo eruption was in 1991. A residence of 6 years in the stratosphere for this exceptional eruption is slightly exaggerated, but we agree that the residence was of few years. The residence time depends on the altitude of the aerosol layer and probably on the nature of the particles; obviously the residence time is shorter in the troposphere. We have changed the sentence to: "their altitude-depending residence time".

L. 23-25 p.9996: It seems difficult to make a correct overview of in situ aerosol measurement without citing Deshler. This place ("aerosol collecting instruments") seems the right one to add some reference to Deshler's work.

The reference is added.

L. 24 p.9996-L. 25 p.9997: I am not sure that using speciation index charts to determine the nature of the particles is very different from choosing a priori hypotheses in the retrieval process.

A priori hypothesis supposes that you know in advance the nature of particles. Using the speciation index is not an a priori hypothesis, since the interpretation of measurements is done using real laboratory reference measurements.

*L. 3 p.9997: The authors could precise that it concerns the **local** size distribution.*

The word "local" is added.

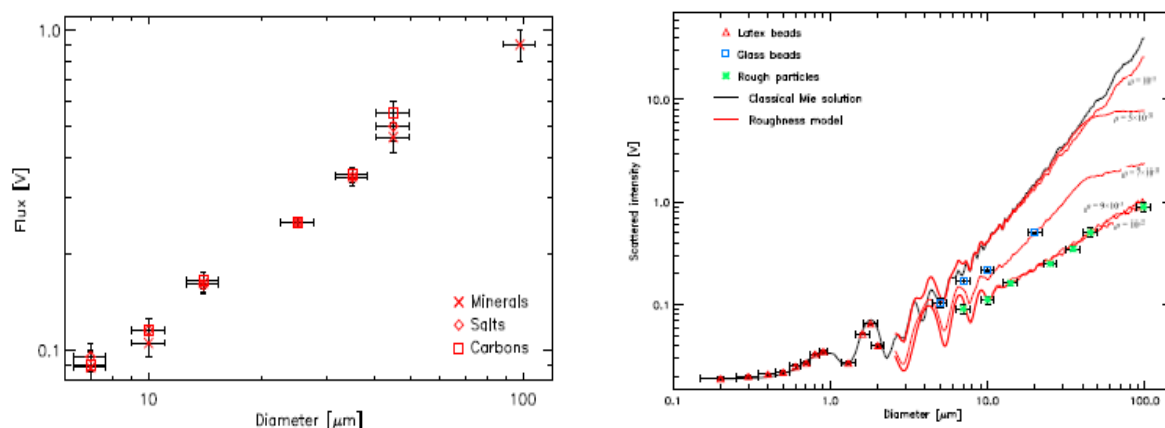
L. 15 p.9997: The authors could add: “[... a given value of their refractive index] and some assumption on their shape”.

This is now added.

L. 26 p.9997, L. 3 p.9998, L. 15 p.10001: Following [Lurton et al., 2014, fig. 8], there is still some dependence in the refractive index. Hence, the authors should use a less strong expression for “such non-dependence”, for instance, “Such **weak** dependence ...”. In the same way, “the light scattered is dependent only on the size” should be replaced, for instance, by: “the scattered light mainly depends on the size...”. And “the 12°-channel, which is insensitive to the refractive index of the particles” in L. 15 p.10001, should be replaced by “the 12°-channel, which is poorly sensitive to the refractive index”.

There is a little confusion on the analysis of the Lurton et al. figures. Figure 5 (below, left) show no dependence for irregular particles. On the other hand, figure 8 (below, right) shows a dependence based not on the refractive index, but on the roughness of the particles.

Following the referee’s suggestions, we have changed the text to: “manly dependent” and “almost insensitive”.



L. 20 p.9998: The concept of “aerosol topology” is not standard, and refers definitely not to transparency properties of the particles. At best, it could refer to the study of the aerosol surface. Do the authors mean “aerosol typology”?

Page 9998 line 20: We have changed “topology” to “typology”.

The scattering properties of particles depend on their size, on their shape and on the real and imaginary parts of their refractive index. Many laboratory studies (e.g. the Amsterdam and Granada light scattering databases, the PROGRA2 database) have shown that the imaginary part of the index, and thus the transparency of particles, strongly affect the light scattered by levitating particles; the effect of the surface is of less importance.

2. Principle of measurements

2.1 Instrument concept:

L. 10-11 p.9999: The fact that the optical chamber is open is obviously not sufficient to say that the pressure is the same as outside. If a pump is used to focus the air flux into a flexible tube toward the optical chamber, many elements let think that the pressure might be highly variable, and definitely often different from outside! This sentence is actually in contradiction with what is mentioned in statement in L. 28-29 p.10000. The description of the device should

be clarified, possibly with a quantification of the importance of the pressure variations, and/or by completing figure 2a.

The reviewer is right. The pressure variations probably change slightly the shape the liquid droplets, removing the Mie oscillations. For solid particles, the pressure changes are not strong enough to modify the shape of the particles. We have removed the sentence on the pressure and the optical chamber.

L. 23 p.9999: The reference is already written in the caption and might be removed to shorten the text.

The reference is now removed.

L. 5 p.10000, L. 25 p.10001, L. 2 p.10002, L. 18 p.10003, L. 1 p.10010: I guess that the “beam homogeneity” refers to the sectional distribution of light intensity. Aren’t there any other issues related to the beam characteristics or stability in time, also possibly related to pressure/temperature conditions (e.g. sensitivity of the laser wavelength to the temperature), and that should be taken into account in the estimation of the uncertainty? LOAC is supposed to measure an extremely broad range of particle sizes, what necessarily requires a large dynamical range. Hence, one can conceive that any sensitivity of the beam intensity to external conditions could affect the dynamical range and introduce a bias in the measurements.

Our technique of measurement is not very sensitive to possible small changes in the laser wavelength, because irregular shaped particles do not produce Mie oscillation.

The intensity of the laser could decrease at very low temperature. We have measured the temperature close to the laser during the stratospheric flights and we are within the nominal temperature operating range. We have added: “The stability of the laser is within $\pm 5\%$; the laser is always operated in is nominal temperature range, even during stratospheric flights.”

L. 14-15 p.10000: I understand that a particle transiting in 500 μs is detected about 20 times (=electronic sampling x transit time)? I guess that size-dependent inertia, or complex motion of irregular grains, etc. may affect the scattered flux. e.g., the small peaks found in Fig. 3 and identified as the contribution of small particles might just be caused by the light scattering by a rotating irregular grain (as indicated by the authors a few lines further). How do the authors estimate the uncertainty on the particle flux due to this kind or effect?

When a (large) rotating particle is outside the beam, it cannot produce the small peaks found before or later its transit inside the beam. On the other hand, the secondary peaks during the transit of irregular grains inside the beam are not considered, since we search for the maximum of the peak only. It is right that the small peaks are produced by small irregular rotating grains. Their mean contribution is retrieved by a statistical approach, when enough grains are detected, as said in the text.

L. 14-15 p.10000: The authors mention a particle transit time “equal or lower than 500 μs ”. But the pulse duration shown in Fig. 3 is about 0.75-1 ms. The authors should adapt their values or explain how to reconcile this difference.

The referee is right, the correct value is 700 μs ; is it now corrected.

Figure 3 shows an example of a 1-ms long pulse corresponding to a 5 μm particle (See text). This allows us to conclude that the flow velocity is about $2 \times 5 \mu\text{m/ms} = 10 \text{ mm/s}$. Using this value and following the same rationale as the authors (L. 10- 19 p.10002), I tried to have some idea on how LOAC is able to detect high concentrations as found by other instruments cited in the paper. I made some intercomparison between the theoretical upper limit of what LOAC is able to detect (supposing a hypothetical population of particles in single file and

infinitely close to each other), and the concentration detected by other instruments. This is the result of these calculations for several plots given in the paper. Corresponding figures and instrument used for the comparison are indicated. I conclude that, in most of these cases, the maximum concentration which can be detected by LOAC is much smaller than the values found by the reference instruments. If I understand correctly, higher values derived from LOAC measurements (using the method given further in the paper) are thus some kind of estimation based on (highly) saturated measurements. Hence, I have some doubts about the reliability of this approach.

We agree that the maximum concentration LOAC can determine is lower than most of the other instruments. But the LOAC limit is far beyond typical concentrations in ambient air. LOAC cannot be used in dense smokes, but can be tentatively used in fog and clouds when the dense concentration correction is applied.

On the other hand, a “large” concentration of particles does not really produce highly saturated measurements. As explained in the text, it is in fact the opposite. This effect can be easily modelled, as explained in section 2.3, and not just “estimated”.

Figure 3: The authors also mention the presence of a few submicronic particles. Are these particles corresponding to very small peaks at time 0.4-0.5, 1.5, 2.3, 2.4 and 3.3 ms? How is the threshold corresponding to the red line determined? Is the quantification of the noise reliable enough to allow the correct detection of real particles producing the weakest signal (1 mV for the smallest particle, following L. 6-8 p.10002), and this for any atmospheric conditions?

The referee is right. We have added in the text: ‘This threshold, or detection limit, is corresponding to the output voltage level on which particles can be detected even if some electronic noise is present. This level is automatically determined by the on-board processor. Because the electronic noise can vary with temperature, this level is determined again every 15 minutes.’

2.2 Calibration:

L. 15 p.10001: I don’t see why the 60° channel doesn’t need to be calibrated: I guess that the flux measured at 60° also depends on the size and the shape of the grains, and that the response is equally affected by the complexity of the particle population, even if the refractive index is the main driver in the measurement.

We agree that our explanations are confusing. We have changed the text to: “The calibration procedure is conducted for the 12°-channel, which is almost insensitive to the refractive index of the particles. The 60°-channel will be used as a comparison to the 12°-channel measurements to determine the typology of the aerosol, as explained in the section 2.4. To conduct such determination, the 60°-channel must have the same output voltage thresholds as the 12°-channel”

L. 4 p.10002: The authors should quantify the expression “low air temperature”.

We have changed the sentence to: The electronic noise is lower than 20 mV at ambient temperature and lower than 10 mV when the electronics are exposed to negative temperatures.

L. 16-18 p.10002: “... size classes [smaller than 0.5 μm] having a width of 0.1 [or 0.2] μm ”: a reference to Table 1 should greatly clarify what the authors mean.

These values refer to usual counting measurements in ambient air, not especially the LOAC ones. We agree that the sentence can be confusing. We have changed the text to: “During

real measurements in the atmosphere, we must ensure that such particle concentrations are indeed present for the LOAC size classes below 1 μm . The LOAC has an integration time of 10 seconds, with a pumping flow of about 2 L per minute. Even in low polluted ambient air at ground (“background conditions”), typical counting measurements available in the literature have shown that concentrations are greater than 1 particle per cm^3 for size classes smaller than 0.5 μm (e.g. Ketzler et al., 2004), which corresponds to $2000/6 =$ more than 300 particles during the 10 seconds LOAC integration time. For particles in the 0.5 – 1 μm size classes, concentrations are greater than 0.1 particle per cm^3 , giving more than 30 particles. Thus, 2 minutes of measurements will provide a good statistics for the LOAC data analysis.”

L. 28 p.10002-L. 3 p.10003: This limitation for stratospheric measurements is important and should be shortly reminded in the conclusions.

We have added in the conclusion: “For lower concentrations, as those encountered in the stratosphere, the data must be integrated during at least 2 minutes to ensure a good statistics of detection”

L. 13 p.10003: The authors should precise “electric flux” in order to avoid the possible confusion with particle flux. L. 16, 21-22, 27 p.10003: The expression “scattered flux”, without more explanation, looks a bit strange. The authors are using the word “flux” possible for 3 quantities: “electric flux”, “particle flux” and “scattered flux”, what can be confusing and requires some caution while using the term “flux”. Concerning the expression “scattered flux”, the authors could move the definition given in L. 22 (“which corresponds to the photodiode output voltage...”) to L. 16. I guess also that this concerns the scattered intensity at 12° only. This should be mentioned.

The referee is right. We have changed the “electric flux” to “output voltage” when speaking of what is recorded by the photodiode; we have changed “scattered flux” to “scattered intensity” when speaking of light intensity, and “flux” to “particles flux” when speaking on air sampling.

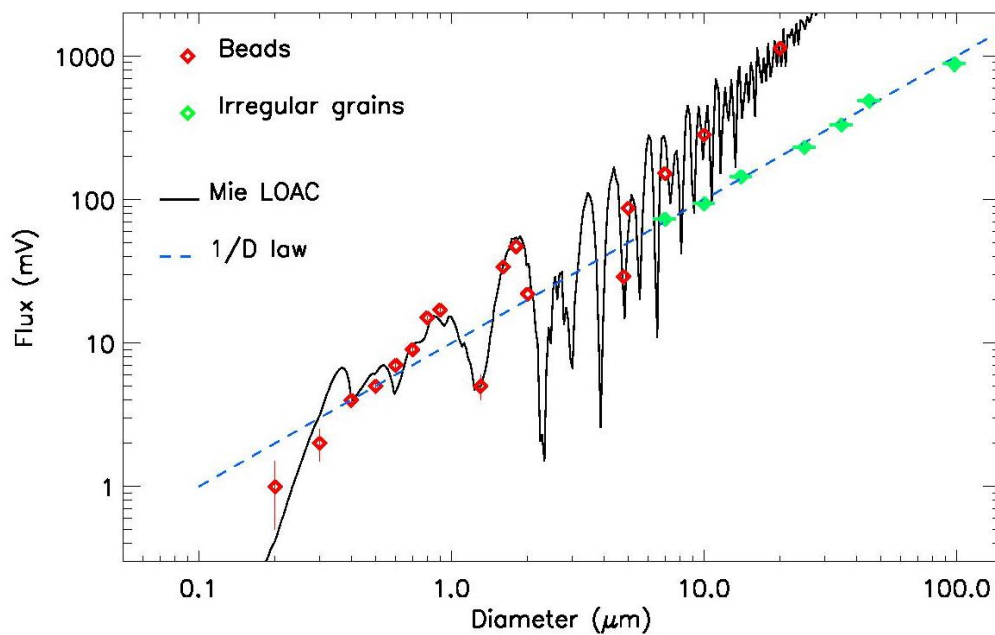
L. 14-17 p.10003: I would like to point out the fact that all scales are logarithmic, what makes relative the expression “no significant dependence”. As an example, following Renard 2010a, there is a factor 1.6 between the reflectance of fluffy silica and sand.

At a given size, the errors bars of the measurements for different natures of grains are overlapping, even in logarithm scale. The Renard et al. 2010 paper shows the 1.6 factor between fluffy silica and sand just for the PM10 measurements. The Lurton et al. 2014 paper presents more recent laboratory measurements, where no significant dependence was observed. Our new calibration with different natures of grains confirms this result.

L. 18-25 p.10003: The formulation of the text is a bit confusing. After explaining the nature of the particles used to make the calibration (this concerns obviously measurements), it is mentioned that an offset is added to some “calculation” of the calibration curve. The authors should explicitly make the distinction between measured and theoretical aspects (where the offset is added) for the clarity of the text.

We agree with the referee that the “offset” effect is unclear. In fact, this offset is the threshold, or detection limit, pretended in Figure 3. We agree that presenting the calibration without subtracting the offset was inappropriate. Thus we have changed the sentence to: “Figure 4 presents the calibration curve for the 12°-channel, with the particle size versus the photodiode output voltage above the detection limit”.

We have also updated the Figure 4, to better present the calibration for the small diameters:



New figure 4

We have added in the text: “Mie theoretical calculations were conducted taking into account the LOAC field of view (11° - 16°). In fact, the LOAC detection of particles smaller than $0.6 \mu\text{m}$ is conducted for output voltage levels where the electronic noise can be not negligible; thus the Mie theoretical calculations must be convoluted with the LOAC noise histogram to be compared to real measurements.”

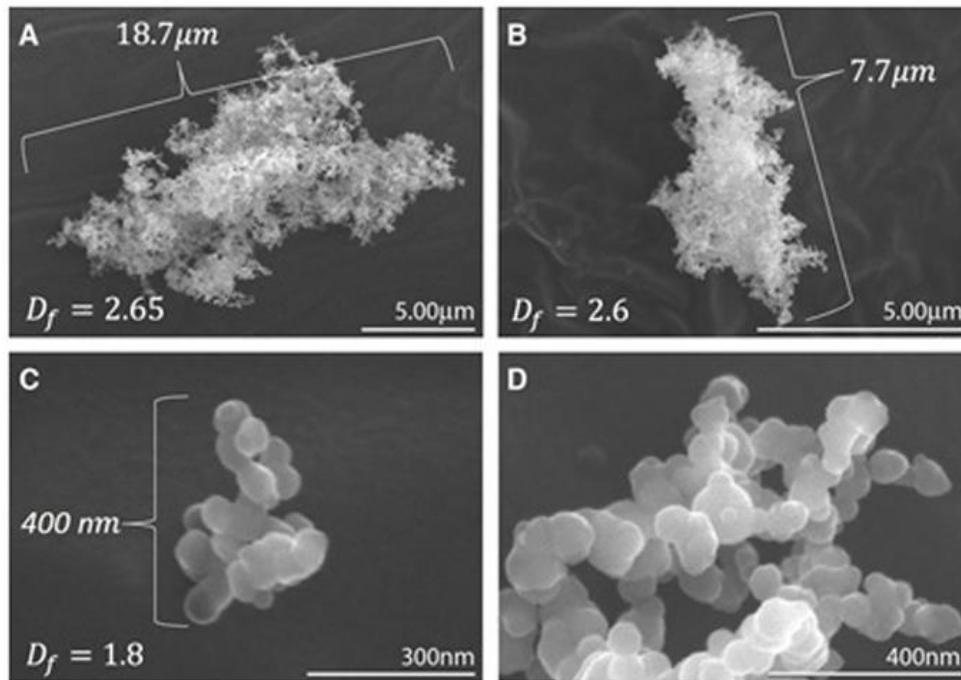
Figure 4: (1) Given as is, this figure raises many questions concerning the experimental validation using 2 kinds of particles (“beads” and “irregular grains”) without any overlap in the size range, and following totally different behaviour. In particular, the discrepancy between the Mie scattering curve and the other response, apparently common to all kinds of possible irregular particles, seems really surprising. Many answers are given in Lurton’s paper, and the authors should explicitly mention that this paper is devoted to an extended study of these aspects. The key-aspects explaining the difference in the behaviour of spherical and irregular grains are only summarized here by a sentence “copied-pasted” from Lurton’s conclusions, and which is in my sense particularly obscure. The authors should revise this explanation to make it understandable by people who didn’t read Lurton’s paper. Joining disparate explanations (L. 25 p.9997-L. 6 p.9998, L. 4-6 p.10004 and maybe others) about the fundamental reason of the choice of the 12° - 16° angle, mentioning Bragg diffraction, and explaining that the “irregular grain” behaviour is obtained even for particles with a very small roughness parameter would really clarify many things.

We agree that we must discuss more in detail the results of the Lurton et al. paper. It is now done in the discussion of Figure 4 (see answer to comment L. 7-13 p.10004). On the other hand we don’t understand why the referee speaks about “Bragg diffractions”, which refers to crystals.

Figure 4 and L. 14-19 p.10004: (2) Despite Lurton’s study, I am not fully convinced by this “calibration”. Amongst the particle types the authors want for sure to measure, we find soot (not considered in Lurton’s paper). Do the authors know the value of the roughness parameter of soot, that possibly looks like aggregates of spherical particles? In view of this particular structure, soot could possibly have a very low value of roughness parameter which

could make its response much more close to the Mie scattering regime. Hence, it could present some oscillatory behaviour, similarly to the glass beads studied by Lurton et al.

Soot particles were presented in the Lurton et al. paper (in fact, they were called “carbon”). A large number of pictures of soot are available in the literature, and they have an irregular shape with surface irregularities; most of the soot are fractal (an aggregate of spherules do not produce a spherical large particle). Thus they have a high roughness parameter value.



Example of wildfires soot, from Chakrabarty et al., *Scientific Reports* 4, Article number: 5508, 2014

Figure 4: (3) In all cases of rough particles considered in Lurton et al., 2014 (even for the most irregular ones), their roughness model (which agrees well with their measurements) shows a range of particle sizes for which the scattered intensity presents an oscillatory behaviour (See figure 8 in their paper). This is of particular importance for the purpose of the present work, where the value of the “flux” is used to determine the diameter of the particles. Unfortunately, these theoretical curves have been omitted here, what masks in my opinion a problem making LOAC probably not effective for this range of particle size (around 3 to 10 μm).

Figure 4: (4) Similarly, for all values of the flux up to about 100 mV, each flux value can correspond to multiple values of the diameter. Below 0.5 μm (range relevant for stratospheric studies), the “flux” is almost constant. Hence, taking into account comments (3) and (4) on this figure, I don’t see how to discriminate unambiguously the particle size for particles smaller than $\sim 10 \mu\text{m}$.

The referee’s interpretation of the Figure 8 of the Lurton et al. is right. It is the limit of the modelling approach, on which some small oscillations are still remaining in the 2 – 10 μm size range. It is why we must validate the choice of using a power law fit to establish a bijective relation between diameter and detector output voltage for the calibration, as shown in the new Figure 4. It is done in the part 3 of the paper, with cross-comparisons of LOAC concentrations and size distributions with those of other instruments. Erroneous discrimination in the LOAC

size particles would produce oscillating size distributions in 2-10 μm range, and inaccurate number and mass concentrations of PM_{2.5} and PM₁₀, which is not the case.

L. 18-21 p.10003: Comments (3-4) on Figure 4 make me conclude that the estimates of the uncertainty on the size determination from the “flux” measurement is much higher than what is mentioned, due to the multiple values of the particle size corresponding to a given “flux” value.

L. 4 p.10004: The concept of “roughness parameter” should be defined. The authors could possibly refer to some work on the subject or using this parameter (e.g. Lurton et al., 2014).

L. 7-13 p.10004: This explanation tends to show that the power law fit should follow much more closely the Mie scattering curve in Figure 4. Actually, Weiss- Wrana’s observations seem to differ significantly from what is observed here (Figure 4), and in Lurton et al., 2014 (their Figure 8). It is not clear to me what the conclusions of the authors are.

We have totally rewritten the discussion of the Figure 4. Here is the new text:

“Mie theoretical calculations were conducted taking into account the LOAC field of view (12° - 16°). In fact, the LOAC detection of particles smaller than 0.6 μm is conducted for output voltage levels where the electronic noise can be not negligible; thus the Mie theoretical calculations must be convoluted with the LOAC noise histogram to be compared to real measurements.

The calibration with the latex beads captures well the large-amplitude Mie oscillations up to 5 μm in diameter. In particular, the amplitude of the oscillations at 1 μm , 2 μm and 5 μm are well reproduced. For the larger sizes, calibrated with irregular grains, the evolution of the scattered intensity (or output voltage) with size is lower than the one expected from the Mie calculation. Lurton et al., (2014), on a paper dedicated to the light scattered at small angles below $\sim 20^\circ$, have shown that, for irregular grains and for a field of view of a few degrees, the scattered intensity could come almost only from diffraction. The authors have introduced in the Mie calculation a roughness parameter ρ , calculated from the standard deviation of the particle shapes from a perfect sphere; ρ is sensitive to the shape of the particles but also to their surface roughness. When ρ is greater than 0.01, the light scattered is dominated by diffraction. Microscopy images of real atmosphere particles greater than a few μm has shown that ρ is always greater than 0.01; as a comparison $\rho \sim 0.005$ for spherical beads. A good illustration of the light scattering properties of such irregular grains can be found in Weiss-Wrana (1983).

In ambient air, the sub-micrometre (sub- μm) particles have also an irregular shape (e.g. Xiong and Friedlander, 2001; McDonald and Biswas, 2004). The Mie oscillations that are present for perfect sphere particles will disappear, being strongly smoothed. The scattered intensity will then increase continuously with increasing size.

The output voltage evolution with diameter for the particles larger than a few μm can be fitted using a power law. The best fit is obtained using a power law in $D^{-1.0}$ where D is the particle diameter. This fit crosses also the middle of the Mie oscillations for the sub- μm sizes, as shown on Figure 4. Thus, it seems reasonable to use this fit for all the particles in the 0.2-100 μm size range, to establish a bijective relation between diameters and detector output voltages. Such fit prevents multiple solutions in the diameter determination for a given output voltage. This calibration approach must be validated by comparison with other instruments and techniques of measurements providing size distribution, which is the purpose of the part 3 of the paper.”

L. 20 p.10004-L. 2 p.10005: Figure 5 does not validate anything, it is just an example of result which may be credible, but is not confirmed nor invalidated by any other data. In view

of the length of the manuscript and of the presentation of other similar curves later in the paper, I would remove this paragraph and the figure.

L. 24-25 p.10004: I don't understand the meaning of this sentence, and more particularly, I don't see what the authors mean with "calibration error": this is probably just an effect of the impossibility to inverse a multi-variate function (i.e. the "calibration curve"), reflecting the limitations of LOAC in the 3-10 μm range (See comment on Figure 4 (3)).

We agree that Figure 5 and the corresponding paragraph are unnecessary. We have remove them.

L. 29 p.10004-L. 2 p.10005: I don't agree on the explanation given by the authors. It seems to me very unlikely that Mie theory fails to describe droplets because they are possibly slightly stretched while passing through the OPC. This would question many decennia of atmospheric research. The use of the fitted power law does bias the size determination. This is clear, e.g. from Figures 10 (See my comments on this figure). It is also very clear from Figure 4 that "flux" measurements by LOAC are not able to discriminate spherical particles with a size below 5 μm . While it is much easier to use the fitted power law for which a flux value always corresponds to one single value of the particle, for particles larger than $\sim 0.6 \mu\text{m}$, the Mie curve shows that this value will generally differ from the real value, possibly by several microns for the largest particles. If the authors pretend to be able to discriminate unambiguously the size of particles supposed to be spherical in the range 0.6 – 10 μm , then they have to provide convincing results.

First, conventional OPCs use a large aperture (tens of degree) and/or white lamp, to remove the Mie oscillation in case of spherical particles. Secondly even if the particles are stretched, the volume is the same, thus the averaged intensity of the light scattered is almost preserved. There is no reason to question previous atmospheric research on liquid particles.

If the liquid particles are stretched, they do not produce Mie oscillations. Liquid particles can be stretched, but not the solid ones when they are injected in the LOAC optical chamber. As shown for measurements inside the clouds and fog, LOAC size distribution does not show oscillations and unrealistic results than can be the consequence of erroneous size attribution.

Since the submission of the paper, we have conducted new cross-comparison measurements with a DMPS and the FIDAS aerosol counter on a sea spray chamber at different temperatures. The instruments have observed first liquid droplets for water temperature above 23° and then salt crystals for lower water temperatures. This is due to the fact that hygroscopic sea salt particles still contain water at the higher temperatures when being sampled ; in other words the particles have not fully effloresced (more details can be found in Salter et al., 2015). The results confirm that LOAC works well even for liquid particles. We propose to add new figures (size distribution comparisons, and integrated concentrations in the 0.2-0.9 μm size range) and a new paragraph to discuss these laboratory measurements at the beginning of the part 3 (see our answer in part 3). We propose to add the name of the two colleagues at the Stockholm University that has developed and operated this chamber (M. Salter and P. Zieger).

L. 3 p.10005: Considering all the comments I made about section 2.2, I can't agree with this statement. It is true that the authors qualify their statement further in the paragraph, but the inability to discriminate particles probably up to 10 μm (See comments on Figure 4), should be mentioned. Further, the sampling cut-off of the inlet discussed later (L. 9-10 p.10005 and mainly section 2.6, in particular L. 27-28 p.10010) could make the detection of very large particles (above a few tens of μm) poorly quantified (See comment on L. 27-28 p.10010). Hence, this conclusion looks really hasardous.

We have given above some new elements concerning the LOAC calibration to better explain the calibration method (the inlet cut-off will be discussed below).

2.3 Concentrations measurements

L. 19-20 p.10005: See remarks on Figure 4 (Section 2.2).

We have provided a new version of the figure 4 and we have improved the discussion (see above). We have also re-organize this section.

*L. 23 p.10003-L. 4 p.10006 : The number of undetected small particles depend on the particle size distribution. The more large particles are found, the higher the amount of undetected small particles. A Monte-Carlo calculation can at best provide “detection efficiency coefficients” giving some indication the number of undetected particles *for the size distribution assumed in the calculation*. But this is only an indication with a reliability decreasing with an increasing number of large particles, and its provides absolutely no information about the real particle size distribution.*

In fact, the Monte-Carlo calculations were conducted for each size class, as already said in the text, not for all the size classes simultaneously. Thus, there is no assumption on the real size distribution. The calculation were conducted for different concentrations of particles in each size class, and similar results were obtained. We have changed the text to: “The ratio of the number of detected particles over the number of injected particles provides the detection efficiency for each size class.”

L. 5 p.10006: I guess that there is no maximum for the transit time! Isn't the peak width increasing linearly with the particle size?!

As a first approximation, the speed of the particle crossing the laser beam is independent of the particle diameter and nature, thus the time transit is always the same (we neglect here the inertia effect).

L. 10-14 p.10006 and Figure 6: How do the author derived this inverse proportionality between real and detected concentrations (i.e. not what is shown in Figure 6!)? I guess that in this extreme case, the system just reaches the saturation, and the detected “flux” tends to keep a giving asymptotically a number of “one particle detected” independently of the real flux. Concerning the correction factor (for particles larger than 1 μm) shown in Figure 6, I guess that statistical fluctuations make the uncertainty on this correction very high.

As said in the text, the curve is obtained by another Monte Carlo approach. The key parameter is that the particles do not cross the laser beam regularly, but randomly. Statistical fluctuations can be minimized by integrated the measurements over several minutes. We have added:” When the mean time between the transit of 2 particles in the laser beam is smaller than the transit-time of one particle in the beam, the detected concentrations became smaller than the real ones, and an inverse proportionality between real and detected concentrations appears”

L. 13-14 p.10007: For the sake of clarity, the authors could make the link between this offset and the “threshold value” in red on Figure 3.

We have removed this paragraph; we have rewritten and moved the “threshold” discussion at the end of part 2.1 (see above).

2.4 Aerosols typology

*L. 25 p.10007 : Do the authors mean “the **scattered light** detected by the 60° channel decreases ...”?*

It is now corrected.

*L. 15 p.10009: I guess the authors mean: "... works well **only** in case of a homogeneous medium".*

We have added "only".

2.5 Reproducibility

L. 11-12 p.10010: The concept of "reproducibility uncertainty" looks strange; the authors should express it in a more standard way.

We agree that the sentence is unclear. We have changed it to: "We must evaluate first the measurement uncertainty of one LOAC copy, and then the reproducibility of measurements from different copies of LOAC in the same ambient air."

L. 13 p.10010: Do the authors mean "a total uncertainty" of $\pm 15\%$? Taking into account all the discussion, I think that the total uncertainty is much more than the sum of these 3 contributions of 5% each. The authors precise "assuming no systematic bias". But I don't think that this assumption can be done, and anyway, this conditions can't be promptly omitted as done by the authors (See e.g. caption Figs 10-13; Paper 2, L. 19, p.10061)

L. 16-17 p.10010: The expression "power law size distribution" is unclear. Do the authors mean "irregular grains for which a power law fit is a valid estimate of the calibration curve"? And if yes, how was the particle size distribution? Like described in L. 4-8 p.10003?

L. 17, 19 p.10010: What do the authors mean by "standard deviation"? Do they refer to the standard deviation of the set of measurements they made in fixed experimental conditions? This is of course very different from the total uncertainty on the concentration, which could be highly biased by the many aspects that raised in Section 2.

Following the referee's questions and comments, we have totally rewritten this part: "The instrument is industrially produced by Environnement-SA (<http://www.environnement-sa.com>); more than 110 copies were produced by the end of 2015. We must first evaluate the measurement uncertainty of one LOAC copy, and then the reproducibility of measurements from different copies of LOAC in the same ambient air.

Tests have been conducted for the different parts of the instrument: diode, pump, photodiode and electronics, to assess the measurements uncertainty that will added to the Poisson counting statistics. The stability of the pump flow over one hour is of about $\pm 5\%$, which induces a $\pm 5\%$ concentration uncertainty. The pump was tested at low temperature and low pressure in balloon flights in the stratosphere and no obvious instability nor loss of performance has been detected. As said before, the laser stability is within $\pm 5\%$. Finally, optical tests have been conducted to evaluate the variability of the response of the photodiodes at given intensity levels. Overall, the detectors response provide an uncertainty of less than $\pm 5\%$ from one copy to another. Taking into account all these uncertainties, we can expect an uncertainty for total concentration measurements better than $\pm 20\%$, for one copy of LOAC.

It is necessary to evaluate the reproducibility of the measurements from different copies of LOAC. In general, the variability of the pump flow was less than ± 0.2 L from one pump to another, but it is recommended monitor the flow rate by a flow-metre before a balloon flight or during ground based measurements; the value of the flow is an input parameter in the post-processing software. Tests have been conducted with 8 LOAC in a "pollution test room" at LPC2E laboratory (Orléans, France). Various types of solid particles have been injected in the chamber. For an integration time of at least 10 minutes, a standard deviation of $\pm 15\%$ (1σ) from the mean concentrations has been obtained between the different instruments for particles

smaller than 10 μm and for the two channels. The standard deviation increases up to $\pm 30\%$ for particles larger than 10 μm , due to the low concentrations of such particles.

The total concentrations uncertainties evaluated for one copy of LOAC and the standard deviation obtained for 8 copies are similar. Thus, we can evaluate that the uncertainty for total concentrations measurements is of $\pm 20\%$ when concentrations are higher than 1 cm^{-3} (for a 10 minutes integration time). For lower concentrations, the uncertainty is dominated by the Poisson counting statistics, up to about $\pm 60\%$ for concentrations smaller than 10^{-2} cm^{-3} . Also, the uncertainty in size calibration is $\pm 0.025\text{ }\mu\text{m}$ for particles smaller than $0.6\text{ }\mu\text{m}$, 5% for particles in the $0.7\text{--}2\text{ }\mu\text{m}$ range, and of 10% for particles greater than $2\text{ }\mu\text{m}$.”

2.6 Inlet sampling efficiency

L. 27-28 p.10010, L. 12-13 p.10011: In an “Application Note” of the firm TSI specialized in precision measurement instruments (Application Note ITI-58, “Measuring total suspended particulates (TSP) with aerosol photometers”, easily available on their website www.tsi.com/uploadedFiles/Site_Root/Products/Literature/Application_Notes/ITI-058.pdf), the sampling efficiency is calculated to a configuration very close to the one used in the paper (TSP inlet, ~30 cm long tube with ~0.6 mm inside diameter, 90° bend and flow rate of 1.7 l/min). Their calculation give penetration efficiencies of 100% for particles in the range 0.1-1 microns, decreasing for larger particles to 75%, 25%, 6%, 1% and 0% respectively for particles of 10, 20, 25, 35 and 50 microns. These values are far less optimistic than the “efficiency close to 100% up to a few tens of μm ” claimed by the authors (L. 28 p.10010). The authors of this note also question the ability of a TSP inlet to provide effective sampling of particles larger than ~20 μm , sizes that seem not considered by the authors in their calculations of the sampling efficiency (cf. L. 27-29 p.10011). Did the authors investigate this issue into detail? Does the use of a beveled metal inlet during balloon flights make the situation more favourable for particles larger than 20 μm .

Yes, we have investigated this issue in detail. In fact, the “TSI” document refers to tube up to a 90° bend. As said in the text, our sampling inlet at ground is vertical, and we have conducted tests on the efficiency of our inlets. For balloon flights, the inlet curve can be slightly bent, and the ascent speed of the balloon taken into account, as explain in the text. We have added at the beginning of part 3: “For all the cases, the inlet is vertical or close to vertical to ensure the best sampling.”

L. 21-26 p.10012: This validation seems very important and should also assess the performances of LOAC in the detection of particles larger than 20 μm .

The modelling work shows that during ascending balloons measurements, the concentrations of largest particles can be overestimated (and not underestimated as suggested by some of the referee’s comments). The analysis is still in progress, but preliminary results seems to indicate that, under these measurement conditions, indeed the concentrations of particles larger than few μm could be overestimated.

3. CROSS-COMPARISON WITH OTHER INSTRUMENTS

We have added a new section on the sea spray chamber measurements:

“3.1 Laboratory concentrations and size distribution (sea spray aerosols)

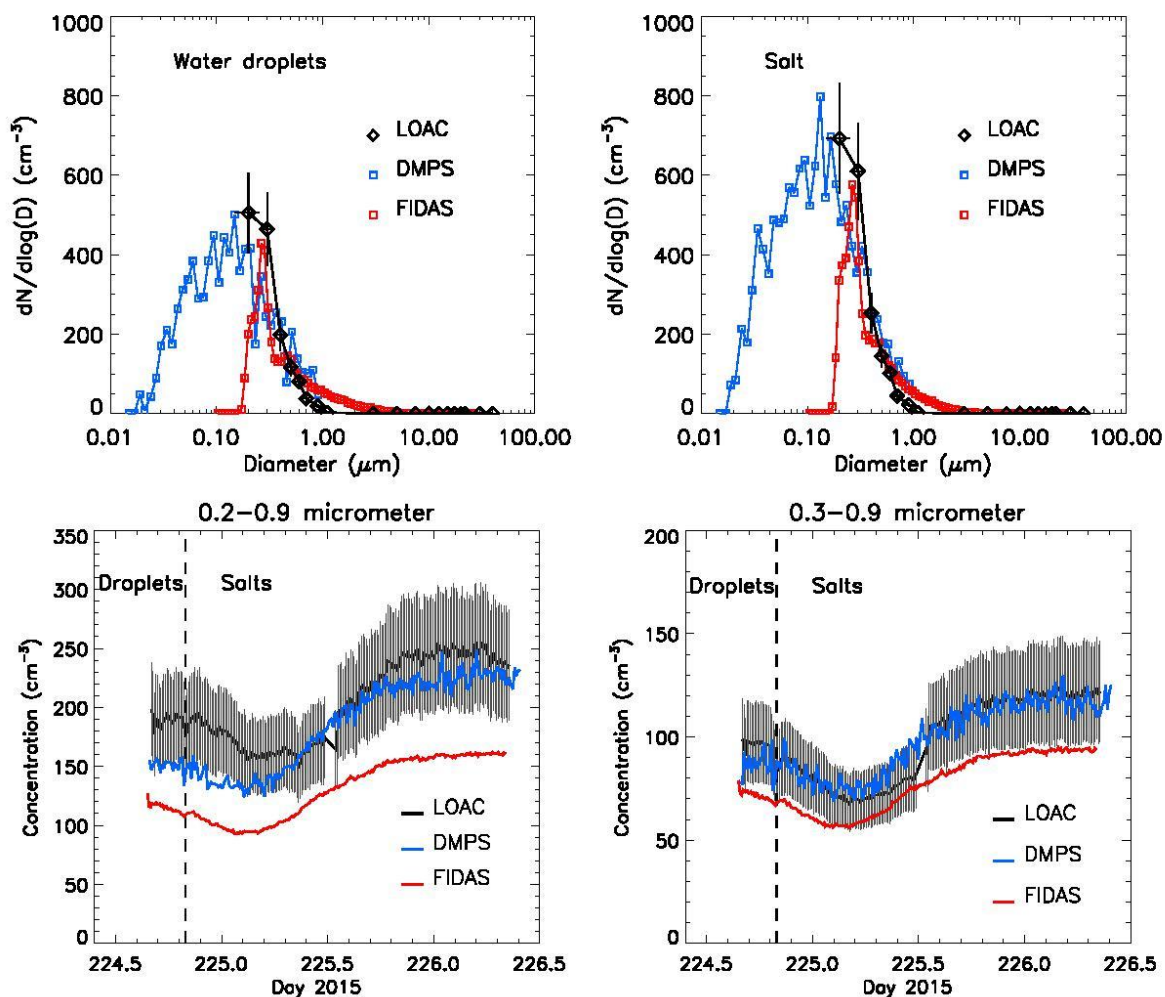


Figure 9: Comparison of LOAC measurements with DMPS and FIDAS measurements performed at the sea spray aerosol simulation chamber at Stockholm University. Top: concentration size distributions for sea spray aerosol particles still containing water (droplets; upper left panel) and at dry or crystalline state (salt; upper right panel). Bottom: integrated number concentration for the 0.2 to 0.9 μm (lower left panel) and 0.3 to 0.9 μm (lower right panel) vs time of the experiment while the water temperature decreased; the transition from seawater droplets to crystalline salt particles (at $T=23^\circ\text{C}$) is indicated as well

A laboratory cross-comparison of LOAC with the FIDAS 200 (Palas GmbH) aerosol counter and a custom built DMPS (Differential Mobility Particle Sizer) (Salter et al., 2014) has been conducted using a temperature-controlled sea spray chamber at Stockholm University, Sweden, from 12 to 14 August 2015. All three instruments were sampling in parallel. The aerosol generation and the air flow were well controlled, thus the instruments have sampled the same air masses.

The sea spray chamber is fabricated from stainless steel components and incorporates temperature control so that the water temperature can be held constant between -1 and 30°C . Air is entrained using a plunging jet that exits a stainless steel nozzle held in a vertical position above the water surface. Water is circulated from the centre of the bottom of the tank back through this nozzle using a peristaltic pump (more technical details on the simulation chamber can be found in Salter et al., 2014). The parameterisation of the sea spray aerosol production as a function of water temperature in the chamber can be found in Salter et al. (2015).

Dry zero-sweep air entered the tank at 8 L min^{-1} after passing through an ultrafilter and an activated carbon filter. Aerosol particle-laden air was sampled through a port in the lid of the sea spray chamber and subsequently passed through a dilution chamber where the aerosols were dried through the addition of dry particle-free air. Following this the aerosol flow was split and transferred under laminar flow to all aerosol instrumentation. To prevent contamination by room air, the sea spray simulator was operated under slight positive pressure by maintaining the sweep air flow several L min^{-1} greater than the sampling rate. Particles produced by the sea spray generation chamber are mainly cubes with rounded edges with dynamic shape factors below those expected for pure cubes.

The measurements were conducted while the water temperature was decreasing. The instruments determined liquid droplets for temperatures above 23°C at the beginning of the measurement session, and then pure salt crystals (dry state) for lower temperatures, as shown by the LOAC typology measurements. This observation is in accordance with the hypothesis of Salter et al. (2015) that the salt particles above 23°C water temperature (which leads to an increased RH in the headspace of the simulation chamber) are not yet fully effloresced and thus still contain water. Figure 9 presents two examples of the size distribution for the three instruments in case of liquid droplets and in case of salts (top), and the time-evolution of the total particle number concentrations in the $0.2 - 0.9$ and $0.3 - 0.9 \mu\text{m}$ range (bottom). The lower limit of LOAC begins at $0.2 \mu\text{m}$ and $0.9 \mu\text{m}$ represents the upper limit of the DMPS. Taking into account the LOAC errors bars, the agreement with the DMPS is very good for the number size distribution and the time-evolution of the total particle number concentration, although LOAC could slightly overestimate the concentration in its first size class. The FIDAS seems to slightly underestimate the concentrations of the sub-micronic particles above $0.3 \mu\text{m}$. The particle size distribution measured by the FIDAS below $0.3 \mu\text{m}$ is strongly influenced by a decrease in the instrument's sensitivity and thus should be generally disregarded. It should be noted that LOAC has well captured the size distribution and total concentration of droplets, which indicates that the assumption concerning the LOAC ability to detect liquid particles is valid."

3.1 Concentrations and size distribution

L. 14 p.10013: If used, the meaning of the acronym "OAG" should be clear at this stage. The authors might possibly refer to the section where it is defined.

The acronym was not necessary here; it is now removed.

L. 6-9 p.10014: The analysis of Figure 10 is particularly short and empty. It is a pity because this figure shows quite well features of the method that are consistent with what is expected from Section 2. For instance, particle class $0.2-0.5 \mu\text{m}$ is very poorly sensitive and fails to detect almost all the particular events or, at least, their amplitude. This is fully consistent with the absence of sensitivity shown by the "calibration curve" in Figure 4. Singularly, some peaks are found by LOAC (e.g. on January 9, 10, 13 and 14) but with the opposite sign with respect to SMPS (minimum peak instead of a maximum or vice versa). Have the authors any explanation for this discrepancy? The same weak sensitivity is found for the class $0.4-40 \mu\text{m}$, although some more events are detected. I guess that this is probably due to the fact that LOAC is more sensitive in the upper part of the $0.4-40 \mu\text{m}$ size range. For particle class $1-50 \mu\text{m}$ (2nd panel), LOAC finds on the contrary a much stronger variability than the fog monitor. Would it be due to a wrong estimation of the size owing to the oscillatory behaviour of the calibration curve found (also for irregular grains) by Lurton et al. (2014) in the range $\sim 0.6-10 \mu\text{m}$? (See also Figure 4 of the present paper and my comments in this figure). The authors

should analyse carefully this figure and the weaknesses it reveals (or rather, it confirms) to really assess what are the exact performances of LOAC, more particularly here for what concerns the sensitivity to size.

Figure 11: Could the strong underestimation of the concentration for the large particles (bigger than $\sim 3\text{-}4\ \mu\text{m}$) with respect to the Fog Monitor confirm a possible strong bias due to the detection blocking in case of large particle (See comments of Figure 3), possibly worsened by aspects of sampling efficiency for large particles (See comments on L. 27-28 p.10010 etc.)? Do the authors have any idea why such a strong underestimation does NOT occur in the case of the fog event illustrated in Figure 13?

Legend Figure 11: concerning the uncertainty, See “General remarks”.

L. 19-22 p.10014: I don't think these are the main reasons to explain the discrepancy. See my comment on Figure 10.

We can understand the referee's concerns. The new figure 9 on laboratory measurements show that LOAC is in good agreement with other instruments. The figure 10 measurements were obtained during “background conditions”, with a relatively low concentration of particles. Some discrepancies could be just due to local fluctuations of aerosols content since the instruments were not so close together. In particular, the LOAC inlet was downward oriented and close to the wall of a building, and can lose the solid particles due to their inertia motion during strong winds, which is not the case during fog events. The referee seems to consider that both the Fog Monitor and the WELAS are reference instruments. The Fog Monitor was calibrated for the detection of liquid particles in fog and cloud; the results in Figures 10 and 11 are just the noise of the instrument and are meaningless. The SMPS measures the electric mobility diameter, which can differ from the optical diameter; it is well known that there are no well-established corrective coefficients between these two diameters. Finally, the WELAS counter, which can be sensitive to the nature of the particles, was calibrated for “mean particles”, and its measurements are unrealistic below $1\ \mu\text{m}$ (as explained in the text). We think that this cross-comparison is quite confusing and redundant with the other cross-comparisons in ambient air, including the new one in the sea spray chamber. Thus we have removed the Figures 10-11.

L. 26 p.10014: Between fog events, LOAC and WELAS disagree most of the time, by up to one order of magnitude!

Yes, but which instrument is wrong? Remember the WELAS problem for submicronic particles. We have added the Rosati et al. reference concerning the WELAS performances: Rosati, B., Wehrle, G., Gysel, M., Zieger, P., Baltensperger, U., and Weingartner, E.: The white-light humidified optical particle spectrometer (WHOPS) – a novel airborne system to characterize aerosol hygroscopicity, *Atmos. Meas. Tech.*, 8, 921-939, doi:10.5194/amt-8-921-2015, 2015.

The same problem was detected with the FIDAS, the successor of WELAS (Pallas Company).

We have rewritten the text for fog event cross-comparison: “Continuous measurements have been conducted in ambient air at the SIRTa observatory (Site Instrumental de Recherche par Télédétection Atmosphérique, <http://sirta.ipsl.fr/>) at Palaiseau, South of Paris, France (48.713°N , 2.208°E), during the ParisFog campaign, <http://parisfog.sirta.fr/>), from November 2012 to April 2013. During this period, the total concentrations of aerosols have been monitored by a WELAS aerosols counter and a Fog Monitor (counter for large droplets)

Strong fog events were observed in November 2012. Particles total concentrations measured by LOAC, WELAS and Fog Monitor are in very good agreement during these events (Figure 10). This result validates the correction procedure applied to the LOAC measurements

in case of dense medium of liquid particles. Figure 11 presents the size distribution at the beginning of a fog event, with the typical enhancement around a diameter of 10 μm (e. g. Singh et al. 2011), and at the end of the event. Both LOAC and WELAS found a bimodal size distribution but disagree for the size and the position of the second mode. Conversely, LOAC and Fog Monitor were in good agreement for the position of the second mode, although the population of the first size class of the Fog Monitor was obviously underestimated. Finally, for the largest sizes, LOAC concentrations are in-between those of the WELAS and the Fog Monitor.

The shape of the size distribution of the WELAS instrument is unusual, as the FIDAS presented above, with a decrease of the sub- μm aerosol concentrations with decreasing size (the opposite trend is expected for background aerosol conditions). The LOAC sub-micronic concentrations measurements are always above those of the WELAS. This could be due to calibration problem of this instrument, as proposed by Heim et al. (2008) and Rosati et al. (2015).

Between the fog events, LOAC and WELAS were sometimes in disagreement. This was due to the difference in the concentration values obtained by the two instruments for the particles smaller than $\sim 0.5 \mu\text{m}$, which may be partly attributed to the WELAS undercounting.”

We have added in the legend of Figure 10: “The peaks of high concentrations correspond to fog events.”

L. 26 p.10015-L.12 p.10016: I think that all what was presented before also reveals clearly identified weaknesses on LOAC’s side in the size range 0-5 μm . The authors should not focus too much on conjectures about what features of other instrument could explain, and just focus on an objective analysis of LOAC’s performances. In particular, there is an obvious problem of sensitivity for LOAC in the range 0.2-0.5 μm confirmed by Figure 10, lower panel. If think that the authors should be satisfied with this kind of explanation, this looks the most evident one.

L. 23-25 p.10015 and Figure 15: I really don’t understand how the authors are able to distinguish the particle classes, and more particularly 0.2-0.3 μm and 0.3-0.5 μm , from the calibration curve. Do they make use of sieves? These figures mainly show the weak sensitivity of LOAC, except maybe in the class 0.5-0.7 μm (thanks the use of sieves?).

As said in the text, the calibration for submicronic size was done using latex beads, not using sieves. We don’t understand why the referee speaks of sieves for the 0.5-0.7 μm range. We disagree with the referee’s comment; the agreement between all the instruments presented above is acceptable or good for particles larger than 0.3 μm , taking into account the errors bars. It must be kept in mind that all these instruments have different inlet systems and pumping, and different calibration procedures. The new version of Figure 4 shows that LOAC can detect small particles.

We have added in the text: “Nevertheless, it appears that the agreements are less good during ambient air measurements than during the sea spray laboratory measurements where the inlets were the same for all the instruments. This is the limit of such cross-comparison in ambient air where the instruments are sensitive to their sampling efficiency.”

Figure 16 and L. 10-12 p.10017: Even in this figure which shows a very good agreement between volume densities computed/observed from LOAC and AERONET, LOAC’s weaknesses are visible: (1) Below ~ 0.5 -1 μm (i.e. 1-2 μm for diameters), the weak sensitivity leads to increased discrepancies between both instruments; (2) For radii ~ 1.5 -5 μm , error increases. This corresponds to diameters of 3-10 μm , for which calibration curve shows (See Lurton et al. 2014) an oscillating behaviour, making ambiguous the determination of the size distribution. This could be mentioned.

The difference the referee points out are for very low volume concentration which are without signification due to the method used for the retrievals. As presented before, there is no error calibration in the 3 -10 μm diameter when considering irregular grains. We say in the text that a small error in calibration (or in size attribution) will strongly affect the volume concentrations, with are proportional to the cube of the diameter. Tests has shown that a 20% errors in LOAC size calibration would shift by several size classes the position of the two modes presented here.

L. 13-17 p.10017: So far, the authors don't present any analysis of the variation of the (randow, systematic) uncertainty with the altitude. Hence, they should not draw any conclusion at this stage on systematic bias.

We have explained earlier in the manuscript that the threshold level, linked to the noise variation with temperature, is checked every 15 minutes during the balloon ascent.

L. 18-22 p.10017: One more time, all these cross-comparisons seem to show in a consistent way, the same weaknesses of LOAC. Several plots present strong evidences of systematic biases between LOAC and other instruments. The authors should revise this paragraph consequently, or just remove it.

“Several” is excessive. We have changed the end of this part to:” This confirms the LOAC calibration and the concentration retrievals are acceptable, at least when the concentrations of the largest particles do not affect the detection of the smallest particles, and when the length of inlet pipe is smaller than few tens of cm.”

3.2 Tropospheric vertical distribution

L. 10-12 p.10018: This is only partly true: For the flight illustrated in the lower panel, LOAC missed an important structure seen by WALL. And in both cases, from ~4 km toward higher altitudes, LOAC's sensivity seems to decrease very rapidly to become fully insensitive above ~5 km. Do the authors see any reason for this behaviour?

Taking into account the errors measurements of the two instruments, LOAC has not really missed any of the important structures detected by WALL. The LOAC extinction were calculated using the concentration for particles greater than 0.2 μm . Since the contribution of the smallest particles is missed, the extinction is underestimated in tropospheric background conditions. On the other hand, in the sand plume, the extinction is dominated by the larger particles, thus the agreement is better (showing that LOAC do not miss the large particles). We have added in the text: “Outside the plume, the LOAC extinction are smaller than the WALL one, because the LOAC extinction are calculated from 0.2 μm , thus missing the contribution of the smallest particles. The extinction presented here must be considered as lower limits.”

3.3 Topology of the particles

Figures 18-22: All these examples seem to demonstrate the effectiveness of the use of LOAC for the determination of the aerosol type. Still, it is interesting to note that, if the “speciation index curves” overall remain within a given “speciation zone”, in the size range corresponding to the smallest particles, these curves show an irregular behaviour with points outside, at the boundary or crossing rapidly the speciation zone of interest. This reflects one more time the lack of sensitivity of LOAC for this kind of small particles, accordingly to the behaviour of the calibration curve (Figure 4).

The referee has made an error. As written in the text, the speciation zones were obtained in laboratory with LOAC, with the same size calibration as for measurements in ambient air. Thus an error in size determination cannot produce some points outside the zones. The discrepancies between real measurements and laboratory zones are only due to the heterogeneity of the studied medium.

3.4 Mass concentrations

L. 14 p.10021: wrong value of the mass density for water.

The microbalance instruments heat the collected samples, to remove the droplets. If we want to compare the LOAC calculated mass concentrations to the microbalance results, we must remove the contribution of the liquid particles. It is why we have put their density to 0. We have change the text to: “A value of 0.0 g/cm^3 was used for water droplets, for comparing LOAC measurements to those of the TEOM instrument, which evaporate condensed water and thus cannot provide mass for water droplets.”

L. 28-29 p.10022: I don't agree with the range of $\sim 0.2\text{-}20 \mu\text{m}$. All the previous intercomparisons confirm a problem of detection for very thin particles, expected from the behaviour of the calibration curve. The reason why this problem does not affect very significantly the mass concentration comparisons, is probably that the relative contribution of mass concentration of thin particles is small with respect to the mass concentration of large particles. This can be verified from the various examples of size distributions given in the paper. The lower limit of $0.2 \mu\text{m}$ is thus not correct.

This referee's comment is in contradiction with the previous ones. If the mass is coming from the large particles, it means that LOAC detected well the particles inside the $3\text{-}10 \mu\text{m}$ range. It is wrong to say that “the relative contribution of mass concentration of thin particles is small with respect to the mass concentration of large particles”. In fact, the size distributions detected by LOAC depend on the nature of the pollution. During the December 2013 event, LOAC shows that at least of 30% of the PM 10 mass and about 50% of the PM 2.5 mass were coming from submicronic particles. During the March 2014 event, most of the mass was coming from particles greater than $1 \mu\text{m}$.

A good example of mass determination is done in the Figure 10 of the second paper.

4. Conclusions

L. 8 p.10023: One more time, submicronic particles should be removed from this range.

Following our previous comments, we maintain our conclusions. We have added:” For lower concentrations, as those encountered in the stratosphere, the data must be integrated during several minutes to ensure a good statistics of detection.

The uncertainty for total concentrations measurements is of $\pm 20\%$ when concentrations are higher than $1 \text{ particle cm}^{-3}$. For lower concentrations, the uncertainty is up to about $\pm 60\%$ for concentrations smaller than $10^{-2} \text{ particle cm}^{-3}$. Also, the uncertainties in size calibration is of $\pm 0.025 \mu\text{m}$ for particles smaller than $0.6 \mu\text{m}$, 5% for particles in the $0.7\text{-}2 \mu\text{m}$ range, and of 10% for particles greater than $2 \mu\text{m}$.

There are some limitations for the concentration retrievals. The measurements of submicronic particles could be underestimated in case of concentration of particles $> 3 \mu\text{m}$ exceeding a few particles cm^{-3} , as encountered in dense clouds or cirrus. Also, LOAC can be sensitive to the sampling conditions. An inlet pipe having a length greater than several tens of

cm can lead to an underestimation of the particle concentration. During flights under meteorological balloons, the retrieved concentrations of the largest particles could be overestimated up to 50% for particles greater than about 2 μm .”

Technical corrections:

L. 4 p.10013, L. 14 p.10001: missing/wrong punctuation.
Not found...

L. 21 p.10002, L. 8 p.10022: incorrect word and symbol.
First one not found; second one corrected.

L. 13-14 p.9998: incorrect sentence.
Not found...

L. 3-6 p.10013: The very long sentence should be splitted.
Correction done.

Figure 10: Typo in the legend: “Fog Monitor”.
This figure is now removed.

Figures 18-23: The authors might consider reducing the figures (e.g. combination left/right panel for Figs. 18-22) to shorten the length of the paper.
We think it will be difficult to read the figure if we combine them.

Reply to referee 3 for the first part of the LOAC paper (AMT-2015-212)

We want to thank the referee for their well detailed analysis that will help us to improve the paper. The referee's comments are in italic and that our replies are in normal style.

General comments

The paper describes a new designed optical particle sizer/counter, the LOAC: the instrument presents a number of characteristics and novelties with respect to traditional instruments which makes it very interesting to be used not only on ground but also under meteorological and stratospheric balloons as well as on unmanned aerial vehicles. Besides that, exploiting and combining the measurements at two scattering angles, the LOAC is committed to be capable not only of retrieving the particles size distribution but also of estimating their nature. The main objective of this paper is to present the instrument and its principle of measurement, and to compare its performance with other conventional instruments to validate it.

The language of the paper is fluent and precise and the overall presentation of the paper is well structured and clear.

The paper addresses relevant scientific questions within the scope of this journal, presenting a novel tool working under the "theoretical" basis of the previous work of Lurton et al. (2014) published in this same journal. However, the authors seem to be very convinced and self-confident of the validity of their instrument and they do not seem very critical in this presentation of the instrument. Given that the instrument presents a number of advantages and innovations, but also some drawbacks, they should provide here a more objective evaluation of the capabilities and limitations of the LOAC. Moreover, they should also present an evaluation of the technical characteristics of the instrument (e.g. final uncertainty resulting from all the uncertainties provided for single aspects of the measurements). The results provided here are not enough to clearly support their confidence that the LOAC does not present any biases. This is of course also reflected in the abstract and in the conclusions of the paper.

The paper presents many figures, some of which could possibly be put in the Supplementary Material or eliminated (e.g. Figure 1).

Overall, the number and quality of references seems almost appropriate, even though some references are missing especially but not only in the Introduction section.

Finally, I would also be very curious to know the answer of the authors to the discussion comment by Dr. Roberts.

We have rewritten the error calculations (part 2.5). We have taken into account Dr. Roberts's analysis (see our answer below). We agree that the conclusion at the end of part 3.2 is too optimistic. Then, we have changed the text to: "This confirms the LOAC calibration and the concentration retrievals are acceptable, at least when the concentrations of the largest particles do not affect the detection of the smallest particles, and when the length of inlet pipe is smaller than few tens of cm."

Specific comments

1. Page 9995, lines 1-24: The abstract should contain not only the advantages and novelties of this instrument, but also its main drawbacks, as well as specific data regarding its uncertainties, detection range and requirements of the environment where it has to be used (for instance, number of particles).

We now provide the measurements size range (0.2 to 100 μm). We have added: “The uncertainty for total concentrations measurements is of $\pm 20\%$ when concentrations are higher than 1 particle cm^{-3} (for a 10 minutes integration time). For lower concentrations, the uncertainty is up to about $\pm 60\%$ for concentrations smaller than 10^{-2} particle cm^{-3} . Also, the uncertainties in size calibration is of $\pm 0.025 \mu\text{m}$ for particles smaller than 0.6 μm , 5% for particles in the 0.7-2 μm range, and of 10% for particles greater than 2 μm . The measurement accuracy of submicronic particles could be reduced in a strongly polluted case where concentration of particles $> 3 \mu\text{m}$ exceeds a few particle cm^{-3} .”

2. Page 9995, lines 23-24: *This sentence is not completely correct, since some disagreements with conventional optical counters are observed. Here it would be better to soften the statement.*

We have removed this sentence, which is not necessary in the abstract.

3. Page 9996, lines 1-13: *In the first paragraph the effect of aerosols on climate is repeated twice, once for its general effects in the atmosphere, and once for its effect in the stratosphere a few lines below. Why? I would suggest reformulating this entire paragraph covering all the different effects of aerosols (health, visibility, climate, ...), and regarding why it is still very important to study aerosols developing new instrumentation, in a more logical way.*

We have removed the second sentence where climate was mentioned.

4. Page 9996, line 15: *This sentence is not correct since the residence time of the aerosols is not always short, depending on various factors such as precipitation most of all; moreover, the residence time of the aerosols is also not constant with height in the atmosphere, with a well-known increasing pattern with height.*

We have changed the sentence to: “...and because of their altitude-dependent residence time”.

5. Page 9996, lines 15-17 and lines 18-19: *Many observation and monitoring systems have already been developed; make some references at least in one of these two sentences.*

The lines 15-19 contain general comments; it seems difficult to provide some references. We provide some references in the following lines, when presenting the main techniques of measurements.

6. Page 9996, lines 22-24: *Since the list of the references is not complete, it would be better to either add “e.g.” at the beginning of the list either extend the list to cover the most important references.*

We have added “e.g.” at the beginning of the references for optical measurements.

7. Page 9997, lines 25-29: *“less sensitive” is not equal to “not-dependent”: I would suggest putting less emphasis on the complete absence of sensitivity in this region in the second sentence.*

We have change “not-dependence” to “low-dependence”.

8. Page 9998, lines 5-6: *You should cite that you will describe the correction for the stray-light contamination offset further on in the paper.*

We have added:” (as explained in part 2.1 and in Renard et al., 2010a).”

9. Page 9999, line 23, and Figure 1: *Figure 1 is actually not needed so it can be put as Supplementary Material or removed.*

We think this figure is important, to show that indeed small ambient particles can be very irregular, and to explain why the optical properties such particle cannot be easily modelled by Mie scattering theory.

10. Page 10000, lines 6-7: *It does not change anything, but if the 55-65° scattering angles is the 60° channel, then for coherence the 12-16° would be the 14° channel...*

The reviewer is right. In fact, there is a mechanical subtlety on the design of the optical chamber. The field of view is 11-16 ° (we have corrected 12° to 11°), but the geometrical centre is at around 12°.

11. Page 10001, lines 15-16: *It is not completely clear in the answer to the referee #1 comments why the 60° channel does not need to be calibrated. This reason has to be better clarified in the text.*

We have changed the text to: “The calibration procedure is conducted for the 12°-channel, which is almost insensitive to the refractive index of the particles. The 60°-channel will be used as a comparison to the 12°-channel measurements to determine the typology of the aerosol, as explained in the section 2.4. To conduct such determination, the 60°-channel must have the same output voltage thresholds as the 12°-channel”.

12. Page 10002, lines 11-13: *How do you ensure that such particle concentrations are present during real measurements? Does the event that such particle concentrations are not present imply any biases or limitations of the use of the instrument?*

The new measurements in the sea spray chamber of the Stockholm University, given on part 3.1 and in Figure 9, were conducted for typical aerosol concentrations in ambient air, with about 200 particles > 0.2 µm per cm³. The concentrations were well below the saturation limit of LOAC.

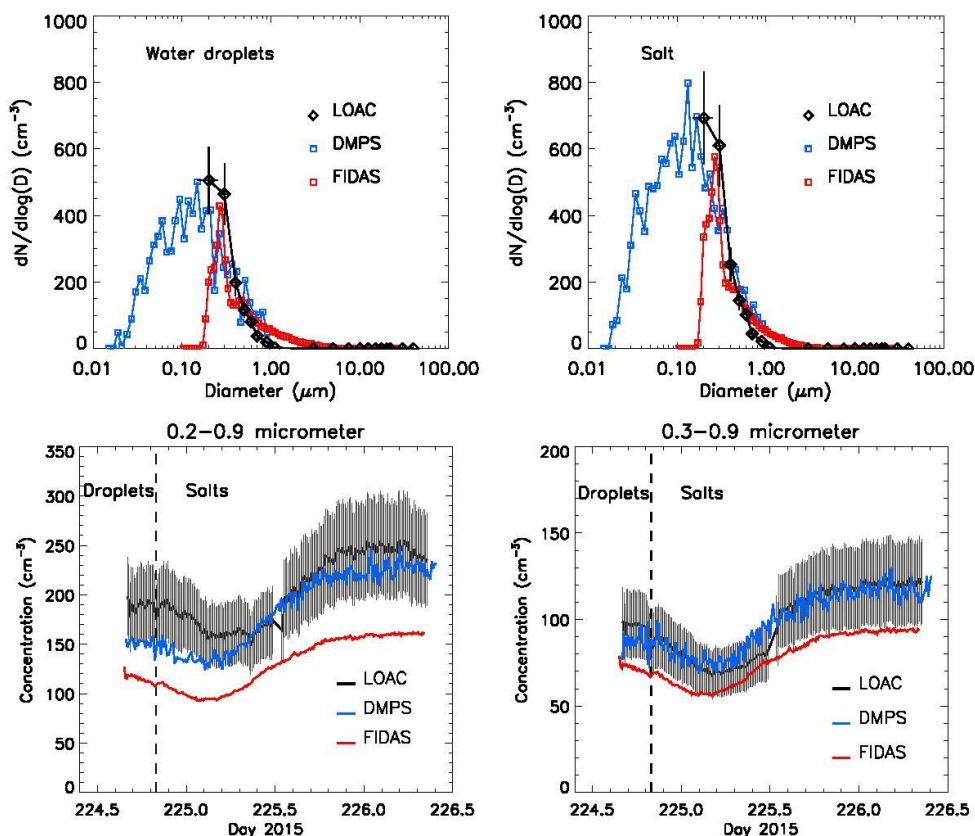


Figure 9: Comparison of LOAC measurements with DMPS and FIDAS measurements performed at the sea spray aerosol simulation chamber at Stockholm University. Top: concentration size distributions for sea spray aerosol particles still containing water (droplets; upper left panel) and at dry or crystalline state (salt; upper right panel). Bottom: integrated number concentration for the 0.2 to 0.9 μm (lower left panel) and 0.3 to 0.9 μm (lower right panel) vs time of the experiment while the water temperature decreased; the transition from seawater droplets to crystalline salt particles (at $T=23^\circ\text{C}$) is indicated as well

13. Page 10002, lines 14-19: Are there any references for these values?

We have added the reference “(e.g. Ketzel et al., 2004)”

Ketzel, M., Wahlin, P. Kristensson, A., Swietlicki, E., Berkowicz, R., Nielsen, O.J., and Palmgren, F.: Particle size distribution and particle mass measurements at urban, near-city and rural level in the Copenhagen area and Southern Sweden, *Atmos. Chem. Phys.*, 4, 281–292, 2004.

14. Page 10004, lines 9-11: This sentence is not clear enough.

We propose to change the text to:” In ambient air, the sub-micrometre (sub- μm) particles have also an irregular shape (e.g. Xiong and Friedlander, 2001; McDonald and Biswas, 2004). The Mie oscillations that are present for perfect spherical particles will disappear, being strongly smoothed. The scattered intensity will then increase continuously with increasing size.”

15. Page 10004, line 27: “In fact” is not correct here since this sentence seems to the previous one.

The paragraph starting page 1004 at line 20 and ending page 1005 at line 2 was unnecessary and a bit confusing. We prefer to remove it.

16. Page 10006, lines 10-11: This sentence is not clear enough.

This sentence was unnecessary here, since explanation is given a few lines below. We have removed the sentence.

17. Page 10011, lines 10-23: It is not clear how the ascent speed of the balloon is taken into account as you answered to referee #1 comments. You report here the sampling efficiency calculations for only one ascending speed.

The ascending speed is in averaged of about 5m/s; it is why we have chosen this value for the calculations. We have added in the text: "Sampling efficiency calculations have been made by considering a mean balloon ascending velocity of 5 m.s⁻¹, which is a typical value when using meteorological balloons..."

We have also conducted new tests on the collecting efficiency. We have added: "Laboratory tests have shown that the LOAC counting can be underestimated when using a collecting pipe longer than about 50 cm, even if the pipe is vertical. Due to the low air flow, some carbonaceous particles become stuck to the walls of the pipe (as shown by the analysis of typology measurements). It is then recommended to use a short collecting system."

18. Page 10012, lines 21-26: This validation is important for the use of the LOAC in many environments where it is committed to be used. I am not convinced this validation can be omitted before presenting (and selling) the instrument to the scientific community, and using it for real measurements.

Such validation must be conducted using many balloon flights, to have a good statistic of large particles measurements. However, the modelling calculations show that the over-evaluation is of the order of 50% or lower, which is similar to the Poisson statistic uncertainty in case of low concentrations. It must be noticed that such study was not conducted for other aerosol counters used in flight. Thus, we propose to say that the errors bars for the larger particles concentrations must be increased. We have added in the text: "Thus, this effect will just affect the retrieved concentrations of the largest particles by about 50% (which is similar to the Poisson statistic uncertainty in case of low concentrations), increasing their error bar."

19. Page 10013-10022: It would be interesting to see the results of the cross-comparison of the LOAC with other instruments not only in terms of time series/profiles, but also in terms of regression lines (maybe to be put in the Supplementary Material).

Since the various counting instruments has not the same size classes, it is very difficult to establish this kind of plot. Also, the regression lines will be statistically biased by the non-homogenous distribution of the concentrations and by the few peaks of high concentrations. It is why we prefer to present time series and dN/dlog comparison plots.

On the other hand, we agree that there are too many time cross-comparisons in the paper. For Figure 21, we propose to replace the time-comparison between LOAC and TEOM to regression line.

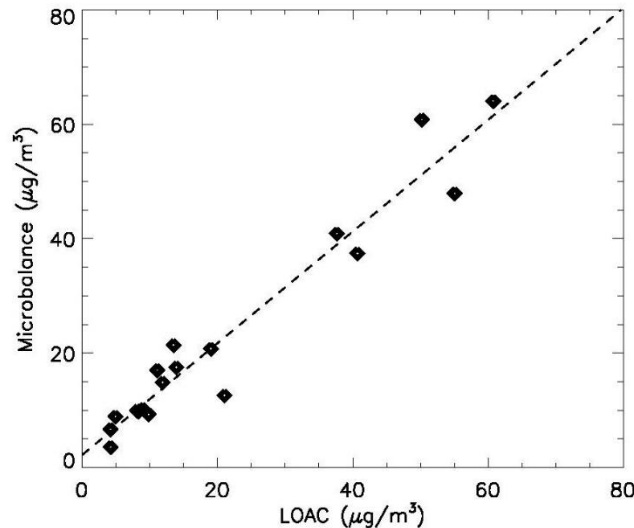


Figure 21: Correlation between LOAC and TEOM microbalance mass concentrations in indoor air (averaged over 24 h); particles have been injected with various concentrations to document a large range of mass concentration

20. Page 10013-100017: There are some other rather famous optical counters which could be used to compare and check the behavior and performance of the LOAC (e.g. Grimm, TSI, ...), possibly not affected by undercounting like the WELAS (page 10014, lines 14-16).

21. Page 10014, lines 27-29: Is it not possible that this disagreement between the LOAC and the WELAS is also due to errors or weaknesses of the LOAC itself? From here it is not possible to be sure that the disagreement is only due to the WELAS undercounting. For this reason it would be perhaps better to compare with an instrument not affected by undercounting.

Now, we present first a comparison of LOAC with 2 other instruments, a DMPS and a FIDAS 200 counter during well controlled aerosol productions and collecting systems, in the laboratory marine sea chamber (part 3.1). Then we present ambient air measurements (part 3.2), which can be more sensitive than laboratory measurement to the inlet and collecting systems. Both FIDAS and WELAS instruments underestimate the concentrations of the submicronic particles. We have removed unnecessary figures for the ParisFog campaign. The comparison with a Grimm instrument is given later in the paper.

We have rewritten this part: “Continuous measurements have been conducted in ambient air at the SIRTa observatory (Site Instrumental de Recherche par Télédétection Atmosphérique, <http://sirta.ipsl.fr/>) at Palaiseau, South of Paris, France (48.713°N, 2.208°E), during the ParisFog campaign, <http://parisfog.sirta.fr/>), from November 2012 to April 2013. During this period, the total concentrations of aerosols have been monitored by a WELAS aerosols counter and a Fog

Strong fog events were observed in November 2012. Particles total concentrations measured by LOAC, WELAS and Fog Monitor are in very good agreement during these events (Figure 10). This result validates the correction procedure applied to the LOAC measurements in case of dense medium of liquid particles. Figure 11 presents the size distribution at the beginning of a fog event, with the typical enhancement around a diameter of 10 µm (e. g. Singh et al. 2011), and at the end of the event. Both LOAC and WELAS found a bimodal size distribution but disagree for the size and the position of the second mode. On the opposite, LOAC and the Fog Monitor were in good agreement for the position of the second mode, although the population of the first size class of the Fog Monitor was obviously underestimated.

Finally, for the largest sizes, LOAC concentrations are in-between those of the WELAS and the Fog Monitor.

The shape of the size distribution of the WELAS instrument is unusual, as the FIDAS presented above, with a decrease of the sub- μm aerosol concentrations with decreasing size (the opposite trend is expected for background aerosol conditions). The LOAC sub-micronic concentrations measurements are always above of the WELAS. This could be due to calibration problem of this instrument, as proposed by Heim et al. (2008) and Rosati et al. (2015).

Between the fog events, LOAC and WELAS were sometimes in disagreement. This was due to the difference in the concentration values obtained by the two instruments for the particles smaller than $\sim 0.5 \mu\text{m}$, which may be attributed to the WELAS undercounting.”

22. Page 10015, lines 20-23: Are there any references to cite for this sentence?

In fact, our recent laboratory measurements have shown that this sentence is not always correct (comment 17.) in case of “long” collecting inlet. We have added: “... which are expected to be not too sensitive to the sampling techniques. Nevertheless, the collecting system used by LOAC was a more than 2-m long pipe, with a risk of losing some (small) carbonaceous particles, as said in part 2.6.”

23. Page 10016, lines 4-5, and Figure 15: Actually the LOAC seems to have missed most of the concentration peaks apart from those in the 0.5-0.7 μm range.

We have added in the text: “Some discrepancies appear for some time periods between the various instruments. For particles greater than $0.3 \mu\text{m}$, LOAC has missed just one peak detected both by the SMPS and the Grimm, at the end of February 3. The peak detected on February 10 by the SMPS were not really detected both by the Grimm and the LOAC instruments (these 2 instruments are in good agreement here). On the opposite, LOAC and Grimm has detected a peak on February 13 for particles greater than $0.5 \mu\text{m}$ which was not observed by the SMPS.

We have added at the end of the discussion: “Nevertheless, it appears that the agreements are less good than during the sea spray laboratory measurements where the air flow and inlets were the same for all the instruments. This is the limit of such cross-comparison in ambient air where the instruments are sensitive to the sampling efficiency.”

24. Page 10017, lines 9-10, and Figure 16: Actually only the volume size distributions are shown.

We say now that it is “volume size distribution”. To reduce the number of figures, it seems no necessary to plot here the dN/DlogD size distribution obtained with LOAC.

25. Figure 16: The 2nd panel actually shows less agreement between the LOAC and AERONET which should be pointed out. Moreover, it is worth to point out here that the AERONET retrieved size distribution result from the Dubovik and Nakajima inversion algorithms (Nakajima et al., 1983, 1996; Dubovik and King, 2000; Dubovik et al., 2000). These retrievals are affected by uncertainties and limitations; in particular fine particles with a mode radius $< 0.05 \mu\text{m}$ for the smallest mode and large particles with a mode radius $> 10 \mu\text{m}$ for the largest mode cannot be retrieved with an acceptable accuracy even in error-free conditions (Reid et al., 2003). The increase of the errors for both cases of very small and very large particles can be explained by the fact that the contribution of these particles to the measured optical characteristics is significantly smaller than for particles of intermediate sizes ($0.1 < r < 7 \mu\text{m}$). Therefore AERONET retrieved size distribution cannot be used as an absolute reference here.

We agree with the referee. As said at the beginning of this part, it is an “indirect evaluation”. We have changed “excellent” to “good”. We have added the Nakajima et al., 1983, 1996 and Dubovik et al., 2000 references. We have changed the text to: “This comparison is just to evaluate the LOAC calibration. Since the volume concentrations are proportional to the cube of the size of the particles, an error in the LOAC calibration would lead to strong discrepancies both in size distribution and volume concentrations, which is not the case.”

26. *Page 10017, lines 18-22: Since there are some disagreements between the instruments in these cross-comparisons, it would probably be better to use less strong sentences here.*

We have changed the text to: “This confirms the LOAC calibration and the concentration retrievals are acceptable, at least when the concentrations of the largest particles do not affect the detection of the smallest particles, and when the length of inlet pipe is smaller than a few tens of cm.”

27. *Page 10018, lines 9-12, and Figure 17: There are some disagreements between the LOAC and the WALI at higher altitudes, possibly due to a lower sensitivity of the LOAC. In addition, you should include your answers to the other referee comments.*

We have added in the text: “Outside the plume, the LOAC extinction are smaller than the WALI one, because the LOAC extinction are calculated from 0.2 μm , thus missing the contribution of the smallest particles. The extinction presented here must be considered as lower limits. Also, the location of the balloon measurements move away from the lidar location as the altitude increases, due to the balloon motion and the wind direction. Thus the discrepancy between the measurements can increase with altitude.”

28. *Page 10018, line 7: Indicate some references for the index.*

We have added the reference: “Wagner, R., Ajtai, T, Kandler, K., Lieke, K., Linke, C., Müller, T., Schnaiter, M., and Vragel, M.: Complex refractive indices of Saharan dust samples at visible and near UV wavelengths: a laboratory study, *Atmos. Chem. Phys.*, 12, 2491–2512, 2012.

29. *Page 10019, lines 28-29, page 10020, lines 1-7: These limitations of the LOAC (among others) should be repeated in the conclusions and perhaps in the abstract.*

We have added in the abstract: “nevertheless, the typology identification cannot be conducted in heterogeneous media”.

It is already said in the conclusion: “LOAC can also provide an estimate of the typology of the particles in case of a relatively homogeneous media.”

30. *Page 10021, line 14: Explain briefly why you used a density of 0.0 g cm⁻³ for water droplets: without explanations, a reader could be astonished for this value.*

We have changed the text to: “A value of 0.0 g/cm³ was used for water droplets, for comparing LOAC measurements to those of the TEOM instrument, which evaporates condensed water and thus cannot provide mass for water droplets.”

31. *Page 10023, lines 4-14: Besides the advantages and novelties of the LOAC, summarize also its main drawbacks and uncertainties.*

We have added in the conclusion: “For lower concentrations, as those encountered in the stratosphere, the data must be integrated during several minutes to ensure a good statistics of detection.

The uncertainty for total concentrations measurements is of $\pm 20\%$ when concentrations are higher than $1 \text{ particle cm}^{-3}$. For lower concentrations, the uncertainty is up to about $\pm 60\%$ for concentrations smaller than $10^{-2} \text{ particle cm}^{-3}$. Also, the uncertainties in size calibration is of $\pm 0.025 \mu\text{m}$ for particles smaller than $0.6 \mu\text{m}$, 5% for particles in the $0.7\text{-}2 \mu\text{m}$ range, and of 10% for particles greater than $2 \mu\text{m}$.

There are some limitations for the concentration retrievals. The measurements of submicronic particles could be underestimated in case of concentration of particles $> 3 \mu\text{m}$ greater than few particle cm^{-3} , as encountered in dense clouds or cirrus. Also, LOAC can be sensitive to the sampling conditions. An inlet pipe having a length greater than several tens of cm can lead to an underestimation of the particle concentration. During flights under meteorological balloons, the retrieved concentrations of the largest particles could be overestimated up to 50% for particles greater than about $2 \mu\text{m}$.”

32. *Figure 10 and 11, captions: Are the LOAC uncertainties indicated total values considering all the aspects (calibration, reproducibility, sampling efficiency)?*

In the legend of figure 10, we have changed 15% to 20% . In the legend of figure 11, we have added: “The LOAC uncertainties are of $\pm 20\%$ for the higher concentration and of $\pm 40\%$ for the lower concentrations.”

In the legend of figure 12, we have added:” The LOAC uncertainties are of $\pm 20\%$ for the higher concentration and of $\pm 60\%$ for the lower concentrations.”

33. *Figure 19, captions: Please insert the coordinates of the location of the sampling.*

We have added in the legend of figure 17 “longitude of $39^\circ 55'$, latitude of $4^\circ 14'$ ”, and in the legend of figure 18 “longitude of $40^\circ 00'$, latitude of $6^\circ 40'$ ”.

Technical corrections

1. *Make sure to change overall along the paper “typology” to “topology”.*

In fact, the correct word is “typology”. It is now corrected in the paper.

2. *Page 10000, line 9: Replace “with” with “of”.*

3. *Page 10000, lines 15-17: Rephrase: “..., a real-time correction is needed for the high stray light contamination at small scattering angles. For this reason, ...”*

The corrections are done.

4. *Page 10001, line 6: Remove “on the”.*

This sentence has been changed.

5. *Page 10002, line 22: Remove “are”.*

6. *Page 10002, line 23: Replace “multiply” with “multiplied”.*

7. *Page 10003, line 15: Replace “with” with “on”.*

8. *Page 10003, line 19: Replace “uncertainties” with “uncertainty”.*

The corrections are done.

9. *Page 10004, line 3: Add “to” before “the”.*

The sentence has been changed.

10. *Page 10006, line 9: Replace “are” with “is” or rather omit the verb.*

The correction is done.

11. Page 10007, lines 8-11: Rephrase: *“For instance, ... Such atmospheric temperature variations can cause a change in the electronic offset with time.”*

The sentence has been changed.

12. Page 10007, line 20: Replace *“they are”* with *“the particles”*.

13. Page 10008, line 7: Delete *“are”*.

14. Page 10008, line 10: Replace *“later”* with *“latter”*. Delete *“will”*.

15. Page 10009, line 19: Rephrase: *“... but the database is still evolving.”*

16. Page 10011: Replace *“long”* with *“length”*.

17. Page 10012, line 16: Move *“has”* before *“always”*.

18. Page 10012, line 23: Replace *“from”* with *“for”*. Replace *“It”* with *“This effect”*. Replace *“could”* with *“should”*.

19. Page 10013, line 4: Replace *“are”* with *“have been”*.

The corrections are done.

20. Page 10013, line 5: Add *“and”* before *“many”*.

The sentence has been changed.

21. Page 10015, line 4: Remove the dot after *“sizes”*. Add *“the two instruments”* before *“disagree”*.

The sentence has been changed.

22. Page 10017, line 11: Missing dot after *“concentrations”*.

The sentence has been changed.

23. Page 10018, line 12: Substitute *“in”* with *“on”*.

24. Page 10020, line 4: Modify *“exhibits”* into *“exhibit”*.

25. Page 10023, line 14: Modify *“from”* with *“obtained on”*.

The corrections are done.

26. Figure 11: *The Figure could be enlarged.*

27. Figure 11, captions: Remove *“good agreement between the instruments”*; *“poor agreement”*; *“The WELAS probably underestimates sub- μm particles (Heim et al., 2008)”*.

This figure, which was not very useful, has been removed.

28. Figure 12: *The Figure could be enlarged.*

This figure is now “Figure 10”. The original size is large; the size was reduced during the AMTD editing process.

29. Figure 13, captions: Remove *“The WELAS probably underestimates sub- μm particles.”*

The correction is done.

30. Figure 14: *The Figure could be enlarged.*

31. Figure 16: *The Figure could be enlarged.*

32. Figure 17: *The Figure could be enlarged.*

All the original figures (or the panel plots) has the same width. Their size was reduced during the editing process.

Reply to T. J Roberts's short comment

We thank Dr. Roberts for the very nice analysis of LOAC measures in “extreme” conditions. She has pointed out a limitation of LOAC use (and perhaps in some others OPC) in case high concentration of large particles. We have analysed recent measurements conducted in dense media as clouds or cirrus, and the same conclusions were derived. Since her work was of high importance for the determination of LOAC accuracy, we propose to include Dr. Roberts as co-author of the paper.

We have added in the introduction: The measurement accuracy of submicronic particles could be reduced in a strongly polluted case where concentration of particles $> 3 \mu\text{m}$ exceeds a few particle cm^{-3} .

We have added at the end of section 2.3: “The LOAC measurements of submicronic particles could be inaccurate in case of concentration of particles $> 3 \mu\text{m}$ greater than few particle cm^{-3} . The high probability of presence of large particles crossing the laser beam will mask the simultaneous presence of the smaller particles; also the response time of the electronics is increased in case of strong illumination of the detectors. These two phenomena will produce a significant underestimation of the concentrations of particles $< 1 \mu\text{m}$. This effect is present in particular in clouds and in fog measurements. For concentrations of particles $> 30 \mu\text{m}$ greater than 1 particles cm^{-3} , as in cirrus, LOAC can miss all the particles smaller than $5 \mu\text{m}$. Thus, concentrations measurements of the smallest size classes in such media must be used cautiously”.