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Interactive comment on “LOAC: a small aerosol optical counter/sizer for ground-based and balloon measurements of the size distribution and nature of atmospheric particles – Part 1: Principle of measurements and instrument evaluation” by J.-B. Renard et al.

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Received and published: 24 March 2015

Answer to reviewer 2 (C210) General comments This paper's goal is to establish the validity of measurements by a new light optical aerosol counter, the LOAC. The authors approach is to use theoretical calculations and Monte Carlo simulations to characterize the instrument, while limiting the presentation of laboratory measurements (Figs. 2 and 3) to a minimum, or to skip them entirely (measurements at the secondary angle and

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speciation measurements) and then, without answering the questions the laboratory measurements raise, to jump presentations of field measurements, and comparisons with other instruments (Figs. 4, 8-22). These comparisons show variations of agreement from good to bad. All of the poor comparisons have explanations, while the good comparisons are used as proof of the LOAC. No error/precision/uncertainty bars are shown on any figure. While instruments have to be characterized first theoretically, they have to also be validated through laboratory measurements. This is where this paper is handicapped to the point that I cannot recommend publication. For the reader interested in the LOAC Fig. 3 is a pivotal figure, and there are a number of serious issues which the authors primarily fail to address, or address with unsubstantiated speculation. After the limited laboratory work is shown the paper transitions immediately to showing atmospheric measurements and comparisons with other instruments. There is no mention of how the instrument is calibrated throughout the paper. While the number of figures showing comparisons is extensive, these are just a handful of examples of a plethora of comparisons. That the authors show, on occasion, a balance of good and poor comparisons is commendable, but there is no attempt to provide summaries of the entire balance of comparisons over a month long field deployment. But such an enterprise is premature. The limited laboratory work presented raises such fundamental questions that it calls into question every subsequent measurement presented, particularly for particles $< 1.0 \mu\text{m}$ diameter. The author's description of the laboratory work, and of a particle's interaction with the light beam as it passes through the optical chamber, is highly speculative and wholly unsubstantiated. Finally there is no description of the calibration procedure, so the reader is left without knowing how the instruments are calibrated. I detail these and many other serious comments on the paper in the following review by line number.

. Most of the answers to the reviewer's concerns on the laboratory calibration can be found in the following paper: Lurton, T., Renard, J.-B., Vignelles, D., Jeannot, M., Akiki, R., Mineau, J.-L. and Tonnelier, T.: Light scattering at small angles by atmospheric irregular particles: modelling and laboratory measurements, *Atmos. Meas. Tech.*, 7, 7,



931-939, 2014, which was indeed cited in the submitted version of the manuscript. Also, a number of the reviewer's comments are erroneous, in particular for the optical properties of irregular particles. It seems that the reviewer wants to promote a method of calibration and measurements developed for spherical particles, and to reject other methods such as ours -mainly developed for irregular grains- without trying to understand how they work.

To limit the length of the manuscript, we have not republished the results presented in the Lurton et al. paper, although it is clearly cited. We understand that some important part of the calibration is missing in the present version of the manuscript, and we will extend the summary of the Lurton paper in the revised version. On the other hand, the reviewer's comments are erroneous concerning the second angle measurements. More than one page is devoted to the concept of speciation measurements and the laboratory calibration. We did not think it was necessary to provide all the laboratory measurements conducted to establish the speciation zones but we could as supplementary material if requested by the editor. The reviewer does not seem to understand the different design of LOAC compared to conventional aerosol optical counters. The calibration procedure he describes works well for instruments performing measurements side scattering measurements (typically at a scattering angle around 90°) that have a large aperture and lenses to collect the light. But LOAC performs measurements at small scattering angles, with a small aperture and without lenses. With such a geometry of measurement, the scattered flux is very sensitive to the Mie oscillations produced by perfect solid spheres and their position inside the laser beam. Thus a new calibration procedure must be developed, as described in the Lurton et al. paper (LOAC cannot work accurately to determine properly the diameter of perfect solid spheres). The instruments used for cross-comparison do not provide errors bars. Thus, it is impossible to plot them. On the other hand, we will add to considerations on LOAC uncertainties in the revised version that LOAC has a $\pm 0.025 \text{ }\mu\text{m}$ uncertainty in the size determination for particles below $1 \text{ }\mu\text{m}$ and of $\pm 10\%$ for particles larger than $1 \text{ }\mu\text{m}$.

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We do not understand the following comment: "While the number of figures showing comparisons is extensive, these are just a handful of examples of a plethora of comparisons. That the authors show, on occasion, a balance of good and poor comparisons is commendable, but there is no attempt to provide summaries of the entire balance of comparisons over a month long field deployment". It seems that we have discussed all the cross-comparison sessions we have presented. Which kind of summary can we provide?

Specific comments:

1210.4-13 and Fig. 2. How is it known that a $5 \mu\text{m}$ particle passed through the instrument and similarly that there were several submicron particles? What sort of deceleration occurs across the laser beam and how is this known? How is it known that secondary pulses are caused by the rotation of the particle in the beam? The entire pulse width is $\Delta t \approx 800 \mu\text{s}$ which seems quite long for such a counter. Is this limited by the electronics or the transit time of the particle across the light beam? This paragraph is highly speculative without any basis as far as the reader can tell. The paragraph should be limited to what is known about the pulse. The authors should know what type of particle is being passed to the instrument and that should be described to the reader. For samples of mono-dispersed particles what does the pulse height distribution look like?

Measurements were conducted in ambient air (we will add this in the legend). The deceleration occurs because the diameter of the optical chamber is larger than the diameter of the inlet. Thus the particles encounter pressure relaxation. All counts statistically above the noise are considered to be real detection. We have observed in laboratory, both by high-speed photodiode measurements and by high speed camera, that irregular-shaped particles exhibit a strong flux variation due to their rotation. We have explained in the text that we consider only the brightest part of their intensity peak. On the other hand, solid spherical particles do not encounter flux variation during their rotation; it is why the LOAC detection algorithm cannot provide accurate results in the

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case of sizing perfect solid spheres. The transit time is due to the width of the laser beam. The following comment is strange: "The paragraph should be limited to what is known about the pulse. The authors should know what type of particle is being passed to the instrument and that should be described to the reader". Of course we know what kind of particles were injected during the calibrations, but we do not know which particles are present in the ambient air? For mono-dispersed particles, assuming that the reviewer speaks of perfect solid spheres, the peaks do not exhibit secondary maxima.

1211.1-2. Irregular particles don't produce oscillations because they have no resonance with the light? What is the basis for this statement? Perhaps they are not called Mie oscillations, which is for spherical particles, but I doubt that means that oscillations are not produced.

Mie oscillations are the consequence of interference in case of particles having symmetries. It is well known that such oscillations disappear for irregular particles, and even for spherical particles with surface irregularities greater than the wavelength (how can you produce interference in this case?): there have been many papers on this subject over the last decades. Our team has been involved in laboratory light scattering by regular and irregular particles since 1993. As an example, please consider the results we have obtained with the goniopolarimeter PROGRA2 and its web data-base www.icare.univ-lille1.fr/progra2/. The following paper also shows a good example of the optical properties of irregular particles: Optical properties of interplanetary dust - Comparison with light scattering by larger meteoritic and terrestrial grains, Weiss-Wrana, K., *Astronomy and Astrophysics* (ISSN 0004-6361), vol. 126, no. 2, Oct. 1983, p. 240-250. We shall refer to those papers in the revised version.

1211.2-4. How is it known that the particles rotate when crossing the laser BEAM?

Rotation of the particles is due to the Magnus force (e. g. 13th European Turbulence Conference (ETC13) IOP Publishing Journal of Physics: Conference Series 318 (2011)

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032021 doi:10.1088/1742-6596/318/3/032021 and reference in this papers). We have observed this effect in laboratory both by high-speed photodiode measurements and by cameras (during the PROGRA2 project).

1211.12-26 and Fig. 3. This figure presents the counter response curve of the instrument and is the essential figure of this paper. Since the counter response curve defines the instrument response to a particle, similar figures have been shown in most papers describing optical particle counters. Typically such curves are presented for comparison against the pulse heights from mono-dispersed aerosol particles to: 1) show that the instrument is well modeled, and 2) to use in calibrating the instrument. This is normally done by adjusting gain stages to match the theoretical response with the practical instrument response for an appropriate particle size. Once the calibration at one size is complete, additional measurements at sizes surrounding the calibration aerosol for that gain stage should fall on the counter response curve, within measurement accuracy, establishing that the instrument is well modeled. I presume that is what is shown in Figure 3 for sizes $< 2 \mu\text{m}$, but the authors do not state so.

This is indeed presented in Lurton et al. cited paper. We will explain it more in detail in the revised version of the paper.

Thus Fig. 3 shows the theoretical counter response using Mie scattering to calculate the response when the scattered light is integrated over the field of view of the LOAC for the detectors at 12 degrees. This curve appears to match the measurements from $0.2 - 2.0 \mu\text{m}$, but then something happens. When larger particles are used, which will cause scattering over the same field of view, the instrument response deviates from the Mie curve as size increases until the difference is a factor of 10 or more. There are a number of issues and questions which arise from Fig. 3. 1) Why is the Mie curve flat between 0.1 and $0.5 \mu\text{m}$? Every other counter response curve, for a forward scattering instrument with which I am aware, shows a decrease in scattered intensity by a factor of 10 or so between 0.1 and $0.5 \mu\text{m}$. 2) Why are there no measurements between 2 and $7 \mu\text{m}$? After the submicron range, this is the next most important part of the size distri-

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bution for tropospheric aerosol. The importance and presence of particles $> 10 \mu\text{m}$ is a lot less certain in ambient aerosol, yet here there are laboratory measurements extending up to $100 \mu\text{m}$. An instrument that cannot provide unambiguous measurements in the size range $0.2 - 7 \mu\text{m}$, will be of limited usefulness for atmospheric aerosol measurements. 3) Why do the measurements deviate from the counter response curve at $7 \mu\text{m}$? The authors suggest that the roughness of the particles leads to this separation; however, they do not support this claim with theoretical calculations or laboratory work. It is possible for the authors to check their statement by generating both rough and spherical particles of large size and to show the variation in pulse height which is claimed between these two cases. Later the authors show results from the LOAC in a cloud and claim that particles $> 10 \mu\text{m}$ are measured. Yet such particles are not rough, so where do they fall compared to the curves in Figure 3? 4) How would T-matrix calculations, which account for non-spherical particles, change the counter response function? 5) How are particles between 0.2 and $0.5 \mu\text{m}$ resolved? The signals from this size range are all around 20 mV . By the time error bars are included, or even without error bars, how can there be any size resolution between these sizes, or even for particles $0.6 - 0.7$ and $1.2 \mu\text{m}$? These results alone call into question all subsequent measurements presented at sizes $< 0.7 \mu\text{m}$, and perhaps even up to $1.2 \mu\text{m}$ once error bars are included. For example, when the instrument returns a pulse of 21 mV , what size is that? Until the authors explain how particles in this size range are resolved, and reconcile that method with Fig. 3, all subsequent results in this size range cannot be accepted, since the reader has no idea how the instrument operates. The standard way to deal with flat regions in counter response curves is to avoid setting any size channels in such a region, or in a region with a double valued response. The same restrictions would apply here. 6) Where are the error bars for the measurements? If they are as large as 50%, which does not seem unreasonable, then at sizes $< 1.7 \mu\text{m}$ the instrument may only be able to distinguish particles of 0.8 and $0.9 \mu\text{m}$ from all other particles sizes below $1.7 \mu\text{m}$. This again raises the question of how the lower size range of particles are distinguished.

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1) We understand the reviewer's concern. In fact, our Mie calculations were converted to voltage (mV) to reproduce the photodiode measurements; we have added to the calculations the offset due to the electronic dark current and high frequency noise of the detector. Then, when sizes decrease, the curve asymptotically decreases to this offset value. We will add this explanation in the revised version of the paper.

2) It is hard to obtain perfect spherical particles greater than a few μm , since surface irregularities can dominate. On the other hand, it is difficult to avoid agglomeration of irregular particles smaller than $\sim 5 \mu\text{m}$. Nevertheless, we have conducted measurements with glass beads of $5\text{--}10 \mu\text{m}$ diameter, as shown in the Lurton et al paper.

3) The reviewer is not correct. We have written: "For the larger sizes, the evolution of the scattered flux with size is lower than the one expected from the Mie calculation. This is due to both the small aperture of the field of view and to the roughness of particles, the recorded flux being dominated by diffraction (Lurton et al., 2014)." Thus, our results on the evolution of light scattered with diameter are supported by these calculations and. Also, we disagree with the reviewer. It is known that droplets encounter deformation in an air flow (see pictures of a moving droplet). Also, the droplets encounter air speed variation in the LOAC optical chamber.

4) You cannot reproduce the optical properties of irregular-shaped particles with T-matrix calculations. Also, you need to know in advance the shape of the particles to conduct such calculations.

5) The reviewer has badly interpreted the figure 3. The scattered flux signal increases monotonously when the diameter increases. There is no double-values responses. Once again, the reviewer makes a confusion between the optical response of spherical particles and irregular ones. In the case of spherical particles, the "flat" response, typically between $0.1\text{--}1 \mu\text{m}$ is just a consequence of the Mie oscillations and of the field of view. In the case of irregular particles, we can statistically classify the irregular-shaped particles in the different size classes. For the $0.2\text{--}0.5 \mu\text{m}$ interval, the pulse

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heights are between 19 and 23 mV, measured with a step of 1mV. As a first approach, assuming a noise of 20 mV, you can statistically have an accuracy of 1 mv assuming 400 counts (square root of 400 divided by 20) or 0.2 mV for 10 000 counts. Several thousands of counts were typically obtained during the 10-second LOAC integration time in ambient air. In fact, the LOAC retrievals are more complex because we must consider the absolute height of the peaks (noise + scattered flux) for each size class, and the histograms of the noise and of the counting.

6) As said above, there is a $\pm 0.025 \text{ }\mu\text{m}$ uncertainty in the size determination for particles below $1 \text{ }\mu\text{m}$ and of $\pm 10\%$ for particles larger than $1 \text{ }\mu\text{m}$. The error for the concentrations is about $\pm 15\%$, on which must be added the Poisson counting statistics. We do not understand where the 50% error hypothesized by the reviewer does come from? In counting? In size determination, in the peak intensities?

1212.1-15 and Fig. 4. This paragraph mixes a discussion of carbon particles and droplets, leaving the reader confused. It is a Paris fog study, so droplets might be expected, and droplets are discussed, and their distortion due to changes in the air flow speed discussed, but again this is speculation for which no evidence is provided. Fig. 4 caption states that it shows carbon particles, but the authors do not show how the measurements in suburban air can be limited to carbon particles, nor how they are capable of resolving the first 4 size bins, when as shown in Figure 3, the signals from these size particles are all nearly identical. There is no indication of the number of measurements made, nor error bars, nor what the overall carbon load is. How in urban air are such particles separated from the rest of the aerosol? How are particles as large as $40 \text{ }\mu\text{m}$ produced in suburban air? Then the authors claim that no bias in size distribution or Mie oscillations were detected for the measurements in fog and clouds. I thought the measurements were for carbon particles, but anyway I doubt the size distribution is so stable that the authors claim is validated and there is no check on it by independent instruments.

As said before, the smallest sizes are resolved. We said in the manuscript that the

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measurements plotted on Figure 4 were integrated over 15 minutes. Since LOAC performs counting every 10 seconds, the number of individual acquisitions used for the figure is $6\text{ min-1} \times 15\text{ min} = 90$. This is very easy to deduce but we shall make the number explicit. We will add errors bars on Figure 4 as suggested. . The total number of particles overall can be found on Figure 21 (10 October is day 287) from AirParif mass concentrations measurements: about $25\text{ }\mu\text{m}/\text{m}^3$. In urban air, particles are mainly carbon, but we agree that other kinds of solid particles can be present. We will remove the word “carbon” and replace it by “absorbing particles”. Obviously, large particles can be present in urban air. The concentration of the $40\text{ }\mu\text{m}$ particles is below $10.5/\text{cm}^3$. This is a very low value, we do not understand why the reviewer is surprised by such a value. The reviewer’s sentence “I doubt the size distribution is so stable that the authors claim is validated” is unclear to us. Regarding comparison with independent instruments, there are indeed comparisons with other instruments in ambient air, as shown in Figures 9, 11 and 12, that the reviewer apparently missed.

1212.24-1213.8. The counting efficiency of an instrument cannot be established with a Monte Carlo simulation. It has to be measured. Particles of known size must be generated and passed to the instrument to be tested along with an independent instrument which measures number concentration. Typically this is done with a differential mobility analyzer (DMA) to generate mono-dispersed particles over a range of sizes, and a condensation nuclei (CN) counter to measure total number concentration. The instrument to be tested is then used to measure concentration over its size range and the concentrations in each channel are compared to the CN counter which will count all particles both above and below any particular size bin. As the size of particles generated approaches and passes each size bin, the counting efficiency should increase and reach approximately 50% at the channel boundary, and then reach 100% at larger sizes. None of this information is provided in this paragraph, nor so far in this paper, yet this is critical information to understand how well the instrument works. The useful range of a DMA extends to $1\text{ }\mu\text{m}$. At sizes up to $1\text{ }\mu\text{m}$ the difference in size between the mobility diameter from the DMA and the optical diameter from the LOAC is in the noise



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of the measurement, or can be characterized for well known aerosol such as ammonium sulfate, or latex spheres. Establishing counting efficiencies at sizes $> 1.0 \mu\text{m}$ is nearly impossible since there is no way to limit a CN counter to only the particles, such as latex spheres, which are generated. All systems to generate such large particles invariably generate a cloud of smaller particles, due to impurities in the generating particle solution, which cannot be separated from the particles of interest since the particle sizes are too large for a DMA. The smaller particles will then be counted by the reference CN counter and confuse the comparison. The technique at larger sizes is typically to use particles which match a bin boundary. The test is then whether the pulses in the channel at the bin boundary matches the counts in the next lower channel, since right at the bin boundary approximately half the pulses will be above the boundary and the other half in the next lower channel, assuming the pulse height distribution will be close to Gaussian as would be expected, and can be measured. The authors state that small particles produce pulses of a few tens of μs , but earlier show a pulse that is near 800 μs and claim that the small particles are thus not detectable in that example. Yet the pulse width depends on the transit time of the particle across the beam not the size of the particle. If there are differences in the transit time that means that the small particles may not generate a signal very far above the noise floor. Establishing whether these signals can be detected is again a matter of generating such particle sizes and showing the counting efficiency, not by modeling it.

“The counting efficiency of an instrument cannot be established with a Monte Carlo simulation. It has to be measured”. It is a strange comment. Why should not model the instrument response? The reviewer argues to use DMA and CN instruments as references. How can one be sure that such instruments provide absolute reference values for concentrations? And how can irregular particles be generated? At most they can be used for cross-comparison, no more. DMA diameters are not optical diameters for irregular particles and there is no simple correspondence. Thus direct comparison cannot be done. The concentrations can significantly differ for measurements of irregular particles, as shown in many papers (e. g. Gulijk, C., Marijnissen, J., Makkee, M.,



and Moulijn, J., The Choice of Instrument (ELPI and/or SMPS) for Diesel Soot Particulate Measurements, SAE Technical Paper 2003-01-0784, 2003, doi:10.4271/2003-01-0784). Also, how can the reviewer postulate that: "At sizes up to $1 \mu\text{m}$ the difference in size between the mobility diameter from the DMA and the optical diameter from the LOAC is in the noise of the measurement"? We have verified with a laboratory instrument that can send the latex beads one by one in the LOAC inlet that indeed LOAC detects all the beads down to 200 nm. We will add this comment in the revised version of the manuscript. But once again, the ambient air aerosol is not made of latex beads, and is it not possible to generate irregular-shaped solid aerosols of mono-dispersed size. We think that an instrument for ambient air measurements cannot just be "calibrated" by latex bead. The only method to validate the measurements of LOAC is to perform cross-comparisons with other instruments using similar and different techniques of measurements. We have presented a long session of inter-comparison with a SMPS, 4 kinds of aerosol counters (Grimm, HHPC6, Welas, Fog Monitor), a photometer, and mass concentrations instruments. We have cross-compared integrated concentration measurements and size-distributions. We believe this is enough.

1213.9-28. Another Monte-Carlo simulation, this time to analyze the issue of coincidence in the beam, or saturation. In fact there are well known methods to calculate ambient concentrations from measured concentrations up to the point of instrument saturation. Beyond that point there is no information in the measurement to allow a true concentration to be derived, since the instrument is in continuous counting mode. This point is reached at a concentration of about 15 cm^{-3} as the authors state and Fig. 5 shows. But what is the point of extending the theoretical calculation beyond that point? The real concentration then becomes double valued. For example, concentrations of 5 and 150 cm^{-3} cannot be distinguished if the measurement is 5 cm^{-3} . This section should only require that the coincidence threshold be analyzed, and that the appropriate parameters to calculate coincidence, and correct for it, be provided. Still it is not clear why the authors make such a clear distinction for coincidence issues above and below $1 \mu\text{m}$. Is this because at smaller sizes the pulses are not far enough above

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the noise threshold to have a pulse large enough to create coincidence problems?

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It is right that: “at smaller sizes the pulses are not far enough above the noise threshold to have a pulse large enough to create coincidence problems”. On the other hand, the reviewer has not really understood how the process of peak detection works with the inhibition / disinhibition system. Additional information, like humidity, is needed to choose between the possible values in case of “saturation”, as said in the manuscript.

1214.8-16. Such temperature sensitivity of the electronic components raises large questions about the usefulness of such an instrument in applications with large temperature swings. Perhaps an electronic recalibration would work, but this is something that cannot be taken on faith, but which must be demonstrated. Merely stating that an electronic calibration is performed without demonstrating its effect is not sufficient.

Words “perhaps” and “faith” are not acceptable in this comment: “Perhaps an electronic recalibration would work, but this is something that cannot be taken on faith, but which must be demonstrated”. It is demonstrated, since all the cross-comparisons were performed in conditions with strong temperature variations (as day-night cycles). Nevertheless, we understand the reviewer’s concern and we will add a sentence explaining these conditions of measurements

Section 2.4 and Fig. 6. Typically instruments that use two angles to estimate particle index of refraction do so by comparing the signal from a single particle. Then the ratio of the pulse heights at the two angles can be shown to be a somewhat unique function of index of refraction. Here the same 10 second integration is applied so the size distributions provided at the two angles will result from a mix of particles with differing indices of refraction. How then is the speciation done? The example that the authors show in Fig. 6 uses particle size resolution between 0.2 and 0.5 μm which show $dN/d\log D$ variation of 100 or more, yet from Fig. 3 there is no indication that such a size range can be resolved. We are not shown a similar curve, or measurements, for the 60 degree scattering angle. Is it similar? If the small sizes are limited by small

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peaks above the noise then we may expect that at 60 degrees the signal above the noise may be vanishingly small. At the larger angle the particles scattering efficiency will decrease. There are ways to quantify the capability of a two angle instrument. Oils with specific indices of refraction are available and can be used in a DMA. Then the aerosol would all be the same. The authors must demonstrate how the LOAC can distinguish such indices of refraction. Without such a demonstration, the reader is left with a paragraph claiming laboratory work establishing the accuracy of the instrument, but without any details. Until such results are presented in the open literature, such claims remain highly questionable and cannot be accepted.

Of course, the speciation does not work well in case of a mix of particles having different refractive indices (as said in the text). The reviewer is wrong when he says: "The example that the authors show in Fig. 6 uses particle size resolution between 0.2 and $0.5 \mu\text{m}$ which show $dN/d\log D$ variation of 100 or more, yet from Fig. 3 there is no indication that such a size range can be resolved". The size ranges are resolved! The 60° channel measurements depends on the refractive index. Once again, why focus on "oils", which are not representative of (solid) ambient particles? The speciation zones presented in Figures 15-19 were derived from curves we have obtained in laboratory with various natures of particles, as said in the manuscript. Of course, we can provide such individual curves if requested by the editor. As said in the text, this procedure works statistically for a large number of detected particles per size class because of their irregular shape, but not for single particles. Again a strange comment: "Until such results are presented in the open literature, such claims remain highly questionable and cannot be accepted". Our results were obtained from long sessions of measurements in laboratory. Of course, if we cannot publish our LOAC results, they cannot be available in the literature! The effect of the refractive index on scattering measurements at around 60° was already published by many authors, including us. As examples, many paper can be found in JQSRT... To summarize, the most important parameter is the imaginary part of the refractive index. Water droplet has value close to 0; mineral (semi-transparent) particles have value in the 10-1-10-2 range; absorbing particles (as

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carbon) has value greater than 0.1.

1216.23-28. The critical piece of information, not mentioned here, is how uniform the laser beam is across the aerosol stream. That the laser flux is quite similar from instrument to instrument is necessary but tells us nothing about the beam quality/uniformity, which is essential if particles of the same size, which pass through different parts of the beam, are to be sized similarly.

We understand the reviewer's concern. The uniformity of the laser is necessary for an accurate counting particle by particle, in the case of spherical (or symmetrical) ones. But once again, for irregular particles and using a statistical approach, the perfect uniformity of the laser is not mandatory; our previous studies for irregular particles (in particular with the PROGRA2 instruments) has shown that the intrinsic flux variation of the rotating particles is higher than the consequence of the non-homogeneity of the laser beam.

Figure 9 and discussion. The agreement with the SMPS at sizes $< 0.5 \mu\text{m}$ is surprisingly good in the top panel, but the question remains, how does LOAC resolve the sizes of particles in the SMPS range when the signals are very similar? From $0.2 - 0.5 \mu\text{m}$, Fig. 3, the pulse heights are between 20 and 22 mV. Why doesn't the SMPS size range extend to near $1.0 \mu\text{m}$ diameter? It should if it is similar to other SMPS instruments.

Figure 9 and discussion. What does the reviewer want to suggest by: "The agreement with the SMPS at sizes $< 0.5 \mu\text{m}$ is surprisingly good in the top panel"? Is he surprised that LOAC can detect particles in the $0.2-0.5 \mu\text{m}$ size range? It is known that SMPS instruments often underestimate the concentrations for particles larger than $1 \mu\text{m}$. We have observed this effect when comparing the SMPS concentrations to those of the other aerosol counters presented here.

1221.7-9 and Figure 10 top panel. Thus in this case the detected concentrations were in the range of $5-10 \text{ cm}^{-3}$ to achieve concentrations of $100-200 \text{ cm}^{-3}$ using Figure 5, correct? How would this correction be done in the absence of an external measurement

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to indicate which side of the curve in Figure 5 should be used? It is also not clear how the correction is applied channel by channel. Figure 10 shows the cumulative concentration from 0.4 – 40 μm . Which channels are saturated? Certainly not all of them.

As said in the text before, the LOAC speciation has indicated “liquid droplets”, thus the “figure 5 correction” procedure is applied. The “saturation” appears for particles larger than 1 μm , for which the time-transit of the larger beam is at its highest value (keep in mind that the small particle saturation is for higher concentrations that were not reached here).

Section 3.2 Speciation. So now, without ever presenting the laboratory data to establish the validity of the speciation determination, the reader is asked to believe that the instrument can fly in somewhat specific, but not exclusive, tropospheric aerosol conditions, in urban air, in dust storms, near the sea surface, in fogs, and in each case make a measurement with a clear identification of that particular species, which was expected, e.g. carbon, sand, salt, water particles. The authors’ figures imply that the tropospheric aerosol is composed of externally mixed particles, of very specific type, throughout the size range from 0.2 – 10 μm , which the LOAC can detect. All graphs are again presented without any error/precision/uncertainty bars on the measurements, or on the lines demarcating the speciation regimes. I was not aware that fogs contained droplets of 0.2 μm diameter and no interstitial aerosol. What is the lifetime of a 0.2 μm droplet? Without seeing the laboratory work that establishes the validity of this technique, nor the uncertainty/precision of the method, nor clear laboratory work confirming the measurements, I find these claims beyond belief. It is certainly out of place for presentation until the prior instrument validation work is published in the refereed literature.

The reviewer has already given this criticism above. The speciation zones include the error bars of the laboratory measurements. The validity of this method was established in the laboratory because the speciation was indeed established in the laboratory! In

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the case of fog, the droplet concentrations are several time higher than background aerosols. In the case of “light” fog or haze, the speciation is not well inside the “liquid droplet zone” but in-between “carbon” and “liquid” zones. We have a large number of measurements showing this situation, and we can provide such figures if necessary.

Section 3.3 Mass concentrations. Based on Figure 3 I can believe that the strength of the LOAC may be at the large particle tail of the size distribution. At least there the signals from separate channels have enough separation in peak height that they may be resolved, so comparisons against a mass measurement may make sense. Thus perhaps these results can be believed; however, I am skeptical based on the lack of rigor so far displayed, and the authors approach to impress the reader with the scale and extensiveness of instrument comparisons, prior to establishing the validity of the measurements.

Section 3.3 Mass concentrations: First, during a strong pollution event mainly due to traffic, a large fraction of the mass is coming from particles smaller than 1 μm ; In this case, erroneous counting of such particles would lead to a mass underestimation of tens of %, which is not the case. The reviewer is contemptuous when saying: “Thus perhaps these results can be believed; however, I am skeptical based on the lack of rigor so far displayed, and the authors approach to impress the reader with the scale and extensiveness of instrument comparisons, prior to establishing the validity of the measurements”. Our results have to be either correct or wrong! It seems that the reviewer refuses to accept that results of LOAC can be good. Our aim is not to “impress the reader” but to conduct an extensive cross-comparison exercise, precisely to establish the validity of the instrument. Is it a lack of rigor to do this? On the opposite, we feel that the reviewer was not rigorous by ignoring the Lurton et al.’s paper that explains the calibration method along with all the works on irregular grains already published, though they were clearly cited in the manuscript as references.

Each of the overall comparisons appears to be part of a larger campaign, so where are the scientific papers using LOAC that should arise from such campaigns. They

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are waiting, I am guessing, on the paper establishing the validity of the measurements, which should be the purpose of this paper. But instead of confining this paper to that primary task, the authors choose to try and do too much, while failing to do the critical work required.

The reviewer seems to make a confusion between calibration and validity. The calibration is mainly presented in the Lurton et al. (2914) paper. Here we focus mainly on the cross-comparison. Calibration refers to the method and samples used to establish the instrumental response. Validation is cross-comparison with others instruments (and not with modelling calculation on ideal particles). Which “critical work required” is missing? According to us, using only spherical particles for calibration, which are not representative of real particles, as done in the past with most of the optical particle counters is critical? We do not feel doing too much by conducting extensive cross-comparisons sessions during field campaign session to evaluate the compatibilities and limitations of LOAC?

Interactive comment on *Atmos. Meas. Tech. Discuss.*, 8, 1203, 2015.

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Fig. 1.