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Atmospheric Measurement Techniques Discussions

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.

Anonymous Referee #2

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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 speciation measurements) and then, without answering the questions the laboratory measurements raise, to jump to

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 μm 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.

1210.4-13 and Fig. 2. How is it known that a 5 μm particle passed through the in-

strument 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 $\sim$ 800 μs 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?

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.

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

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 μm, but the authors do not state so.

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 μm, 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 μm? 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 μm.

2) Why are there no measurements between 2 and 7 μm? After the submicron range, this is the next most important part of the size distribution for tropospheric aerosol. The importance and presence of particles > 10 μm is a lot less certain in ambient aerosol, yet here there are laboratory measurements extending up to 100 μm. An instrument that cannot provide unambiguous measurements in the size range 0.2 – 7 μm, will be of limited usefulness for atmospheric aerosol measurements.

3) Why do the measurements deviate from the counter response curve at 7 μm? 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 μm 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 μm 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 can be any size resolution between these sizes, or even for particles 0.6 – 0.7 and 1.2 μm? These results alone call into question all subsequent measurements presented at sizes < 0.7 μm, and perhaps even up to 1.2 μm 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 μm the instrument may only be able to distinguish particles of 0.8 and 0.9 μm from all other particles sizes below 1.7 μm. This again raises the question of how the lower size range of particles are distinguished.

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 μm 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.

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 μm. At sizes up to 1 μm 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, or can be characterized for well know aerosol such as ammonium sulfate, or latex spheres.

Establishing counting efficiencies at sizes > 1.0 μm 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.

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 μm. Is this because at smaller sizes the pulses are not far enough above the noise threshold to have a pulse large enough to create coincidence problems?

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.

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/dlogD 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 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.

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.

Figure 9 and discussion. The agreement with the SMPS at sizes < 0.5 μm 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 μm, Fig. 3, the pulse heights are between 20 and 22 mV. Why doesn't the SMPS size range extend to near 1.0 μm diameter? It should if it is similar to other SMPS instruments.

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

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.

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.

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 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.

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