Answer to reviewer 1 (C227)

We thank the reviewer for their his comments that will help us to improve the manuscript.

General comment:

In order to evaluate the instrument the authors present a vast amount of atmospheric measurement with aerosol conditions which can not be considered well constrained. From the perspective of the reader it is almost impossible to get an idea of the capabilities and limitation of the instrument.

We don't understand what the reviewer means by : *"In order to evaluate the instrument the authors present a vast amount of atmospheric measurement with aerosol conditions which can not be considered well constrained"*.

It is true that aerosol conditions during atmospheric field measurements are not constrained. We have tried to present comparisons with other instruments for a range of well-documented conditions:

- Urban background ambient air
- Fog conditions (confirmed by other instruments)
- Saharan sand (confirmed by other instruments and by satellite data)
- Sea salt aerosols

According to us such cross-comparisons in field conditions present a good way to validate the instrument performance under actual atmospheric conditions.

We have also produced well controlled conditions in the laboratory of pure samples of carbon, sand and salts, used to establish the reference values for the speciation index.

Detailed comments:

1)) *The authors use the index of refraction to explain the concept of the speciation. In the following text they use the vague phrase "nature of the particles" as if there is more to it then the index of refraction. What is it and what is its expected effect on the instruments functionality?*

The reviewer is right. There is some confusion between "index of refraction" and "nature of the particles" in the text. Indeed, the speciation is sensitive to the refractive index of the particles. We have tentatively referred to it as the nature of the particles. Perhaps "type" is better than "nature". We will change this in the revised version of the text, and clearly define our terminology.

2) *To better explain the concept of speciation the authors should consider plotting mie curves for the two angles as a function of refractive index.*

We do not use Mie scattering for the calibration of the instrument. To better understand why, please refer to the paper cited in our manuscript: 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, 931-939, 2014. We will add an extended summary of the Lurton et al. paper in the revised version.

The speciation concept is based on empirical laboratory measurements. Obviously, the Mie calculations can show the well-known effect of the refractive index on the scattered flux, but they could induce confusion into our discussion since we do not use such calculations.

3) *Figure 2: a Particle with a size of 300 nm would result in a peak 3 orders of magnitude smaller than a particle with a diameter of 5000 nm. The 5000 nm particle in figure 2 has a amplitude of 80 which means a 300 nm particle would give rise to a peak with an amplitude of 0.08 which is well below the noise level in figure 2. With such signal to noise ratio the smallest detectable particles would have a diameter of ∼ 700 nm.*

We agree with the reviewer's comment about figure 2 under the condition that one considers perfect (transparent) spheres and Mie calculations typically at large scattering angles, with a large aperture and a lens to collect the scattered flux across tens of degrees. But LOAC uses an unusual design for the light scattering measurements: measurements at small scattering angle, with a small aperture (few degrees), and no lens.

At small scattering angles and with a small aperture of a few degrees, the scattered flux is very sensitive to the position of the spherical particles inside the laser beam, and can vary by a factor of 10 or more. This is why LOAC cannot be used to accurately detect small solid perfectly spherical particles. On the other hand, the situation is totally different for irregular particles (see answer to point 5).

4) *Page 1210 line 5 and Figure 2: The authors never mention which of the two detectors this data is recorded with.*

The reviewer is right. It is for the 12° channel. We will add this in the revised version.

5) *Page 1211 line 1: "Fortunately, real atmospheric particles are not perfect spheres and will not produce Mie oscillations." This sentence is misleading and wrong.*

We totally disagree with the reviewer. Real atmospheric particles are not perfect spherical spheres (with perhaps few exceptions like metallic spheres). In fact, we must replace "perfect spherical sphere" by "solid perfect spherical sphere". Some particles have a compact irregular shape, other ones have more or less a fractal shape. Thus, the Mie scattering calculations are not valid. Even more sophisticated models (T-Matrix, DDA…) need to know in advance the global shape of the particles to calculate the scattered flux. Even liquid particles are not perfectly spherical during the LOAC measurements, since they are slightly distorted by the speed variation of the air flow in the collecting system and in the optical chamber.

The Lurton et al. paper shows the effect of irregular shaped particles on the scattered flux at small scattering angles when the measurements are performed with a small aperture. In this case, we are mainly sensitive to the diffraction.

For the case of irregular particles, the flux scattered by the particles crossing the laser beam is not monotonous, as shown in Figure 2. This is a consequence of the particle rotation, due to the Magnus force. We have observed this well-known effect in the laboratory (both by high-speed photodiode measurements and by high speed camera). We have explained in the text that we consider only the brightest part of the intensity peak, which cannot be directly related to theoretical calculations.

6) *The mie calculation in figure 3 is incorrect. It looks to me as if the theoretical data is not actually plotted in log scale along the y axes. In a loglog plot the slope should be ∼6 for diameters smaller than the laser wavelength.*

We understand the reviewer's concern. Our (correct) Mie calculations were converted to voltage (mV) to reproduce the photodiode measurements, and we have added to the calculations the offset due to the electronic dark current and high frequency noise of the detector. The curve asymptotically decreases with decreasing size to this offset value. This was explained in the Lurton et al. paper. We will add this explanation in the revised version of the manuscript.

7) *I do not understand the author's argumentation of the origin of the upper count limits (page 1213). The width of a detected peak is, unless the size of the laser focus is diffraction limited, dominated by the dimensions of the laser. This means the actual duration of a scattering event and therefore the width of a peak is the same for all particle sizes and so is the probability for coincidences. My suspicion is that the instrument needs coincidences in case of small particles in order to get the scattering intensities above the detection threshold.*

We agree that our text could be confusing in this matter. We think that the reviewer speaks about the detection of particles smaller than 1 μm. When the signal of the scattered flux is just above the noise, we cannot detect the whole transit of the particles inside the laser beam. We just detect the brighter part of the peak. Thus the apparent transit time of the particles inside the beam is shorter than for the larger ones. Instead of waiting 500 μs to start a new counting, the apparent transit-time can be reduced down to 30 μs. Taking into account the detection efficiency for the smaller particles, up to 3000 particles per cm^3 can be (statistically) detected. We will improve the discussion in the manuscript to make this clear.

8) *To get a better picture of the accuracy and precision of the instrument the authors should show a figure of the size distributions when sampling the calibration material,e.g. PSL at 200 nm, 400 nm and 800 nm and some for the calibration material produces with the sieves.*

This was presented in detail in the Lurton et al. paper. What do we de for the present manuscript?

9) *The authors estimate an uncertainty in the count rate of small diameter particles of 15 % purely on the statistical deviation between different instruments and state that LOAC has no systematic bias. However, when looking at the comparison between different instruments there are strong deviation which can not be explained. In particular in the 200- 300 nm regime I see no correlation to other instruments.*

The uncertainty we end up with is ±15%, leading to 30% uncertainty. Also, there is also an uncertainty in the size determination, of ±0.025 μm for particles below 1 μm, and of ±10% for particles larger than 1 μm. We will add these values to the revised version of the manuscript.

We agree that we have assumed no systematic bias, which could be optimistic. On the other hand, the other instruments we have used for the cross-comparison do not provide errors bars. This makes it difficult to evaluate any possible systematic bias in the LOAC measurements.

The cross-comparison for the 0.2-0.3 μm size-bin is conducted with an SMPS instrument, which is based on the electric mobility diameter of the particles. This diameter can strongly differ from the optical diameter used by LOAC. In particular, fractal particles have a larger surface than compact particles, which can bias the estimation of both the electric mobility diameter and the optical diameter. Thus the retrieved concentration in a given size range can strongly differ between the two instruments. Since the gradient in concentration as a function of particle size of the size distribution could be high for particles greater than typically 0.1 μm, a discrepancy of a few hundredths of μm in the size determination can produce up to a factor 2 in the retrieved concentrations.

On the other hand, the 30 % uncertainty of the LOAC concentrations is in agreement with the mean discrepancy with other optical counters presented on Figure 13.

10) *Some technical details which would be useful for the reader: a) Instrument dimensions b) Do you need some additional computer to run the instrument or is all the peak analysis, data storage, etc. done on the board visible in fig. 1 c) Does the instrument capable of transmitting its data? d) what is the diameter of the sample stream? e) how does the instrument adjust to pressure changes without affecting the flowrate? f) Is there a sheath flow? g) how does the instrument manage to measure light intensities over so many orders of magnitude? (the scattering intensity ratio between 200 nm and 100 um diameter particles is theoretically ~ 6 orders of magnitude.)*

Some technical details are given in the 2nd manuscript. But we agree, we need to provide somewhere (perhaps in the 2nd manuscript) more information.

a) The size of the optical chamber is about 20 x 10 x 5 cm^3.

b) The analysis of the peaks is performed on the board in Figure 1, and provides every 10 seconds the concentrations detected by the 2 channels. An additional computer is necessary to record and analyse the data.

c) The LOAC can be used under different conditions. When deployed underneath meteorological and tropospheric balloons, the data are transmitted in real time by telemetry. For deployments under large stratospheric balloons, the data are stored on-board using a specific module. On the OAG tethered balloon, the data are sent to the ground using a Wi-Fi link and are stored on a computer. An autonomous version for automatic ground-based applications is now also available, including an on-board computer to record the data.

d) About 2 x 4 mm^2.

e) Tests during a flight by meteorological balloon have shown that the rotation speed of the pump (vane-type one) is constant for all pressures encountered.

f) There is no sheath flow.

e) We have answered above.