Reply to referee 2 for the second part of the LOAC paper

Overall this is a clearly presented paper describing a range of initial results provided by a newly developed instrument, LOAC, designed to measure aerosol size and type, which has great potential to provide novel results. The paper achieves the goal of describing a range of new measurements, though some of the measurements, specifically the aerosol speciation index, need to be more clearly explained, and uncertainties in the observations should receive more attention. This should be easily achieved by modifications to the article. Although there are a lot of plots in this paper, they are all relevant to the topic of the article and straightforward to navigate through.

We thank the referee for his useful comments that will help us improving our paper.

Speciation Index Plots

The first time the reader comes across data relating to the speciation index is p10066 L2, yet the authors have not explained what this data actually means or shows, other than a brief mention at the start of section 2. For example, what do the range of numbers (such as in Fig 9) ranging from 0.1 to 4 mean? How do these separate different aerosol types? What conclusions can be taken from the figures when the black data points fall in zones covered by more than one aerosol type? The black data points/line should be included in the figure captions. What is the uncertainty in the speciation index observations?

Although the authors provide a brief overview of how the instrument works and observations provided at the start of section 2, it would be useful to expand this a little more to summarize the main findings of part 1 paper, and in particular expand on the speciation index as described above.

This comment is in contradiction with the referee 1 comments. We have added that a description of the instrument can be found in part 1 of the paper. Nevertheless, we agree that the description of the speciation index is too short.

We have changed the description of the speciation index to: "The measurements at 12° are almost insensitive to the refractive index of the aerosol particles and are used to determine the concentrations. On the other hand, the measurements at 60° are very sensitive to the refractive index. A "speciation index" is retrieved by combining the 12° and 60° channels measurements. This index is mainly sensitive to the imaginary part of the index, thus to the absorbing properties of the particles. Speciation zones have been determined with LOAC from numerous in laboratory for 4 families of particles: solid carbonaceous, mineral dust, and salt particles, and liquid droplet. The carbon speciation zone is larger than the other ones because the scattered light properties of such particle depends also on their shape and their composition (organics vs. soot). The speciation indices obtained from LOAC observations in the atmosphere are compared to the speciation zones obtained in the laboratory to derive the typology of dominant particles in different size classes."

We have added in the typology figure captions: "the LOAC data are black dots"

Minor Comments

10061 L17-18 – It would make more sense to report these values as, "up to _3000 particles cm-3 smaller than 1 um" and the same for "20 particles cm-3 greater than 1 um" rather than the order of wording as used currently.

It is now corrected.

10063 L6 – do the authors mean, "ejected from" instead of "rejected inside"?

We confirm that the aerosols are rejected inside the gondola during balloon flights. For long-term ground-based measurements at a fixed point, the aerosols are rejected outside the box that contains the LOAC. We have changed the text to: "For all kinds of free short flights, the measurements were not conducted in the balloon wake, preventing contamination."

10064 L20 – should be 'submicron'?

We usually speak of "submicronic" particles, in opposition to "micronic" particles.

10064 L23 – "4 times smaller at 220m" – this is not clear on the plot due to the log scale used. In general, the log scales used on figs 3, 5, 8, 13, 15 and 17 are useful for showing differences of large magnitudes, such as between the different size bins and for order of magnitude changes of aerosol loading in individual size bins, but not changes of the order of say a factor of 4, as shown here. Are the authors able to provide additional plots, perhaps as a supplement, which better illustrate these changes?

The changes are now more obvious even in log-scale after re-scaling the figures.

10065 L1 – is the inlet operated when the instrument is flown on the UAVs as well?

We have added in part 3.2: "The sampling on UAV is performed by a vertical inlet, in a non-turbulent zone of the UAV environment.", and in part 3.3 for the OAG balloon:" inlet vertically oriented".

10065 L21 – replace 'on the opposite' with 'contrastingly'

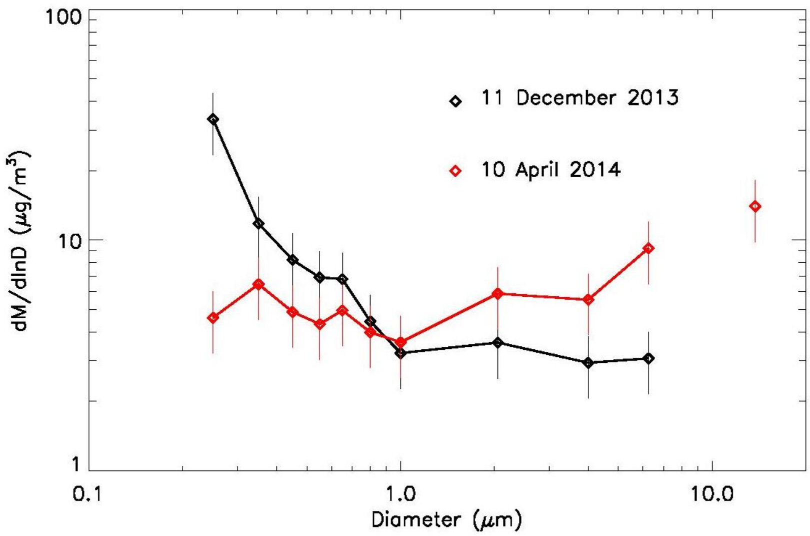
The change is done.

10065 L27 – remove 'between'

The change is done.

10065 L28-29, 10066 L1 – it is not clear from the size distributions showing dN/dlnD that the pollution mass is dominated by the smallest particles, since the larger particles, although fewer in number, will contribute more to the mass. Please include a plot of dV/dlnD or dM/dlnD overlaying the dN/dlnD plots to demonstrate that this is the case.

The reviewer is right. We have added a new figure showing a comparison of mass measurements (dM/dlogD) for the 2 flights considered here. We have added: "The size distribution at 3 different altitudes (Figure 9) shows that the pollution is dominated by the smallest particles. Also, the mass concentrations (calculation method is presented in the paper 1) are dominated by the small particles, in comparison with the 10 April 2014 measurements (Figure10)."



“Figure 10: Comparison of the mass concentration distribution for the 11 December 2013 strong pollution event and the low pollution conditions on 10 April 2014; the measurements are at ground.”

Section 3.4 – it would be useful to add ‘with drifting balloons’ to the title for this section to help keep the reader informed with which measurement operation was used in each section/campaign described throughout the paper.

The change is done.

Section 3.4 – are drifting balloons retrieved? If so, how is this done logistically?

Unfortunately not. Because these balloons were not allowed to fly above continents, they were automatically forced to land on sea at ~30 km from the coast. Some of the gondolas were recovered and sent back to us but contact with seawater did not allow us to reuse the material.

10066 L26 - and following ambient wind directions and speeds, presumably? If the ambient dust/air layer is slowly descending, will the balloon also descend with the layer, or remain at a particular altitude?

The balloon will descend with the layer. We have added in the text:”*, following ambient wind directions and speeds.*”

10067 L10 – change ‘been’ to ‘be’

Change done.

10067 L24 – change ‘save up’ to ‘conserve’

Change done.

Figure 12 – a colour bar should be included

The colour bar table was lost during the figure processing. This is now corrected.

Figure 13 – please use a more useful time for the x-axis, such as time of day, hour of day.

The change is done.

Figure 13, and other plots showing dN/dlnD – what are the uncertainties in these values? It would be useful to show these on the plots.

The uncertainties were removed for clarity reasons. We have added in the legend: "the uncertainty (at 1σ) is of about ± 20% for concentrations higher than 10^{-1} cm^{-3} up to ± 60% for concentrations lower than 10^{-2} cm^{-3}".

Figure 13 – Can the authors comment on the changes in dN/dlnD in the largest sized particles shown in the plot and any reason for these changes? Or are the variations within the uncertainty of the measurements?

We have added in the text: "The variation of concentrations of these particles are coming from the Poisson statistics, and from natural variations in case of very low concentrations. A good estimate of their mean concentrations can be obtained by averaging (or integrating) the measurements over a longer time period."

Fig 13 – as stated above, it's difficult to see small changes in size distribution in the plots due to the log scale used. It would be useful to include a size distribution plot, say contrasting a few points in time from the measurements, such as at the start and end of the data shown in fig 13.

We have used the log-scale to be able to present all the size class concentrations. We think that the global time-evolution can be seen on this plot. We prefer to maintain this figure as it is, since our purpose was not to detect small changes in size distributions.

10068 L9-12 – '..suggesting no significant sedimentation of large particles during the flight or compensation by particles from above' – or it is possible that deposition to the layers below was equally compensated for by deposition from the layers above. If the layer is slowly descending, will the balloon follow this descent?

It is what we meant to say by "compensation by particles from above", but it could be unclear. We have changed the sentence to: "a compensation of particles sedimenting in the layer below by particles sedimentation from layers above".

The balloon will follow slow synoptic movement of the air mass but not particle sedimentation by gravitation.

10068 L12-14 – these results of large particles being retained during mid-long range transport are consistent with those found by Ryder et al. (2013), Weinzierl et al. (2011) and Denjean et al (2015) who reported similar measurements, and should be cited.

We are not sure that our results can be directly compared to the result of Ryder et al. and Weinzierl et al., where most of the large particles were deposited in less than one day. Also, the Denjean et al. results stopped at 30 µm. We propose to add this text:" The shape of the average size distribution is consistent with other counting measurements perform from aircraft (Ryder et al., 2013; Weinzierl et al., 2011; Denjean et al., 2015), although the concentrations can be different because the measurements were not conducted in the same plume events.

10069 L9 – how do the authors know the particles are sulphuric?

It has been well known for a long time that liquid stratospheric aerosols are a mixture of water and sulfuric acid. In fact, LOAC has detected liquid particles; it is not possible to say more about their composition. We have removed the word "sulfuric".

10069 L11 – see comments above about order of notation (change to 'below 1 particle cm-3 greater than 0.2 um').

Change done

10070 L4-7 – for what reason are mineral dust particles originating from the surface ruled out?

To our knowledge, it seems impossible to lift to the stratosphere terrestrial micronic-sized particles, expect of course during strong volcanic eruptions. Not such eruption has occurred since the Mt Pinatubo in 1991.

10072 L15-20 – Nicoll et al. (2011) also performed observations of charge in dust layers – this publication should be cited here.

Indeed, this paper is very important. We have added: "Also, Nicoll et al. (2010) has reported that charge is present within layers of Saharan dust, from 2 balloon soundings performed above Cape Verde Islands."

General questions about LOAC – it would be useful if explanations to these questions could be covered somewhere in the manuscript:

Is the instrument affected by passing through cloud? Do clouds need to be avoided?

LOAC has passed through clouds several times, and was not affected by them. Nevertheless, LOAC has crossed one time a cumulonimbus cloud and the balloon was destroyed. We have added in the text:" LOAC has passed through clouds several time and was not affected by them, except once when the balloon was destroyed inside a cumulonimbus. In case of measurements inside cirrus clouds, the concentrations of the smallest sizes can be underestimated due an optical shadows effect, as explained in the part 2.3 of the first paper."

Could the instrument detect/isolate biological aerosol particles or pollen particles?

In principle, yes, since the 12° channel can detect all kinds of aerosols, but this has not been checked yet. New laboratory measurements must be conducted with such particles to determine their speciation zones and to see if they significantly differ or not from the other zones.

It seems that the instrument would be useful in detecting dust particles at high altitudes in the atmosphere which may act as ice nuclei particles, which are normally notoriously difficult to measure. Can the authors comment on the instruments applicability for this?

LOAC could probably distinguish these dust particles from other aerosols, using typology results. Preliminary analysis of the flights seems to indicate that solid particles can be sometimes distinguish from liquid particles for the smaller sizes, but this needs confirmation.