Reply to referee 1 for the second part of the LOAC paper

We want to thank the referee for his comments that will help us to improve our paper. Nevertheless, we disagree with by some of his comments.

Please note that the referee's comments are in italic and that our reply in normal style.

General Comments

As first sight, this paper gives the impression that it only brings very limited amount of new results from experiments of which part of them are already described in Paper 1. Many repetitions of information contained in Paper 1 let rise questions about the usefulness of this article. The authors don't hesitate to use copy/paste, what is particularly irritating, distracting and not really respectful for the reader who might spend his/her time in a more useful way.

The quality of the text should definitely follow more closely the standards of a quality scientific journal.

All these repetitions occupy about a third of the length of the paper, which is, taking account the need to read Paper 1 to have an overview of the LOAC principle, much too long. Most of the photographs bring, in my opinion, very few real added value, if any. The one in Figure 1could be useful but, unfortunately, no element gives any idea of the real dimension, missing an important part of its usefulness.

Section 2 also brings very few new elements in the description of the instrument.
In Section 3, the report given about measurements at "Observatoire Atmosphérique Generali" includes once again repetition of informative background from Paper 1, again sometimes just "copied-pasted" from it. Results from this campaign are spread over Paper 1 and 2, while a synthesis of them would make them much more readable, interesting and pertinent for such a review of the LOAC project.

Concerning the scientific approach, its looks like the only objective pursued by the authors is to show that "it flies and seems to work in all conditions". A detailed analysis of the data is never really provided. There is no attempt to validate seriously the results, nor even to try, if reference data are really missing, to investigate the effect of changing experimental conditions (e.g. different weather conditions for a same location) on the measurements. An important test would be to see the impact of the choice of inlet on the response, e.g. on the detection of large particles. But this can't be assessed from this work, and the choice of inlet is even not always provided.

I also regret the poor quality of the manuscript, which seems written very fast and still presents many typos.

We disagree with the fact that we have copied/pasted parts of the paper 1. It is not true that there are repetitions of paper 1 that occupy about 1/3 of the paper. A careful reading of the two texts shows that no more than a few tens of lines are in common, and they concern only the description of the instrument and some conditions of measurements. Also, it is not true that they are repetitions of paper 1 that occupy about 1/3 of the paper.

We believe that all the photographs are necessary to show the different LOAC gondolas. We agree that the dimensions were missing. We have added in the paper" its size is 28 x 18 x 15 cm^3."

Also, it is not true that part 3 presents copied-pasted results from paper 1. In paper 1, we have presented mass concentrations derived from LOAC measurements at "Observatoire Atmospheric Generali" and comparison with normative "Airparif" measurements. Observations were performed in different weather condition, including storms. On paper 2, we present a vertical profile of particle concentrations and the associated typology. This is totally different.

The cross-comparisons of LOAC in different geophysical conditions were already presented in paper 1; note that we prefer to speak of "cross-comparison" than of "validation" since there are no absolute reference instruments for particle counting. Tests on the response of the different inlets has been conducted in laboratory. We confirm that no large particles are lost with the inlets used here. Also, we have presented in paper 1 theoretical calculation in case of flight under meteorological balloons.

We discuss here the results from two tethered balloons measurements (Vienna and Paris). These data are totally new. Also, we present and discuss new results from drifting balloons inside a desert dust plume, which is also totally new, as well as the particles concentrations and typologies measured from meteorological balloon flights. About 6 pages are devoted to the description and the analysis of these new results.

We agree that information concerning the inlet is not provided here, although the information are available in paper 1. We have added in part 2:" The inlet is almost vertically oriented towards the sky" for the meteorological balloons, and "In this case, the inlet was horizontally oriented, to avoid the contamination from the balloon" for the drifting balloons. We have added in part 3:"vertically oriented" for the OAG tethered balloon campaign.

Specific comments:

Abstract

Remarks given in the review of Paper 1 should be taken into account, more particularly remarks about the expected limitations and real performances over the size range targetted by LOAC.

Since we have presented in the revised version of paper 1 some new results confirming the performances of LOAC, we prefer not changing the text here.

2. LOAC instrument and gondola for balloon flights

L. 19 p.10061: Concerning the estimate of the uncertainties, see comments in the referee review of Paper 1.

We have improved the discussion on uncertainties in the revised version of paper 1. We have added in the text: "For a 10-minutes integration time, the uncertainty (at 1σ) is of about ± 20% for concentrations higher than 10^{-1} particles cm^{-3} up to ± 60% for concentrations lower than 10^{-2} cm^{-3}; the uncertainty for total concentrations measurements is better than ± 20%. Also, the uncertainties in size calibration is of ± 0.025 μm 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."

L. 4 p.10062: Inconsistency with Paper 1, where a weight of 350 g is mentioned.

The referee is right. We have changed to (in the 2 papers) "300g."

3.1 General comments

L. 6 p.10063: What do the authors mean by: "the aerosols were rejected inside the gondola"? One should expect that aerosol are rejected "outside" to avoid the creation of a pollution cloud.

Rejecting aerosol outside the gondola could create a local pollution. Since the inlet is outside the gondola, rejecting the aerosols inside the gondola is safer to prevent a contamination. Also, the measurements are not conducted inside the balloon wake. We understand that this sentence can be confused, and we propose to change it to: "For all kinds of free short flights, the measurements were not conducted in the balloon wake, preventing contamination."

3.2 Unmanned Aerial Vehicle flights

This campaign is not indicated in Table 1, and there is no mention of the kind of inlet used for this campaign.

Table 1 is for balloon flights as said in the legend. The UAV flight was not conducted during a campaign; it was a test flight. We have added in the text: "The sampling is performed by a vertical inlet, in a non-turbulent zone of the UAV environment."

3.3 Tethered balloons

Campaign in Vienna: The authors just give some profile of vertical size distribution (Figure 5) with an analysis of the aerosol topology. However, a lot of efforts were made to improve the air quality in Vienna, and this city disposes on a dense air quality measurement network, monitoring continuously the air of the city. Hence, I guess that a lot of data are available to validate LOAC's measurements, or at least to make some first intercomparison between size distributions measured in the city. Why isn't it done?

Air quality measurements on cities are performed at ground by microbalances (see Paper 1 for the Paris air quality measurements), to provide masse concentration of PM_{10} (particulate matter smaller than 10 μm) and sometimes of PM2.5. No information are available on the size distribution and on the nature of the particles, and their evolutions with altitude. Thus no cross-comparison can be done.

Figure 4: I am not sure that all these pictures really bring significant additional information to the discussion.

It was the first time that such a small tethered balloon was operated with an aerosol counter that sends the data in real time by telemetry, and that provides an estimate of the nature of the aerosols. According to us, the figure contributes showing that LOAC can be easily deployed for such kind of measurements.

Campaign OAG: This long-duration campaign provides most probably enough data to illustrate all what is mentioned in the text. The authors could show the contamination due to construction activities, the difference between size distributions (as a function of the size) at ground level and at some selected interesting altitudes, during events of well-mixed air and during pollution events. Episode with a visible accumulation layer could be shown with the temperature profile to visualize the temperature inversion and the effect on the layer. The *authors, clearly, miss many opportunities of interesting discussions, and again, no validation is proposed using local air quality measurements.*

It is interesting to note that no particles are detected with a size higher than 20 μm. Could it be due to the characteristics of the TSP inlet (See referee report on Paper 1, comment on L. 27-28 p.10010, L. 12-13 p.10011)?

The aim of the paper was not to present all the results obtained during the OAG campaign; it will be done in an up-coming paper. The construction activities are in fact out of the scope of the present paper. We propose to remove the sentence (we can discuss on these data with the referee if he/she is interested by these measurements). But, as suggested by the referee, we have added a new vertical profile in case of low pollution and well mixed air, and we have changed the text to: "Most of the time, the air was well mixed and the concentrations were almost constant with increasing altitude for particles smaller than ~10 μm. Figure 8 (top) presents an example of such situation during a low level of pollution, on 10 April 2014."

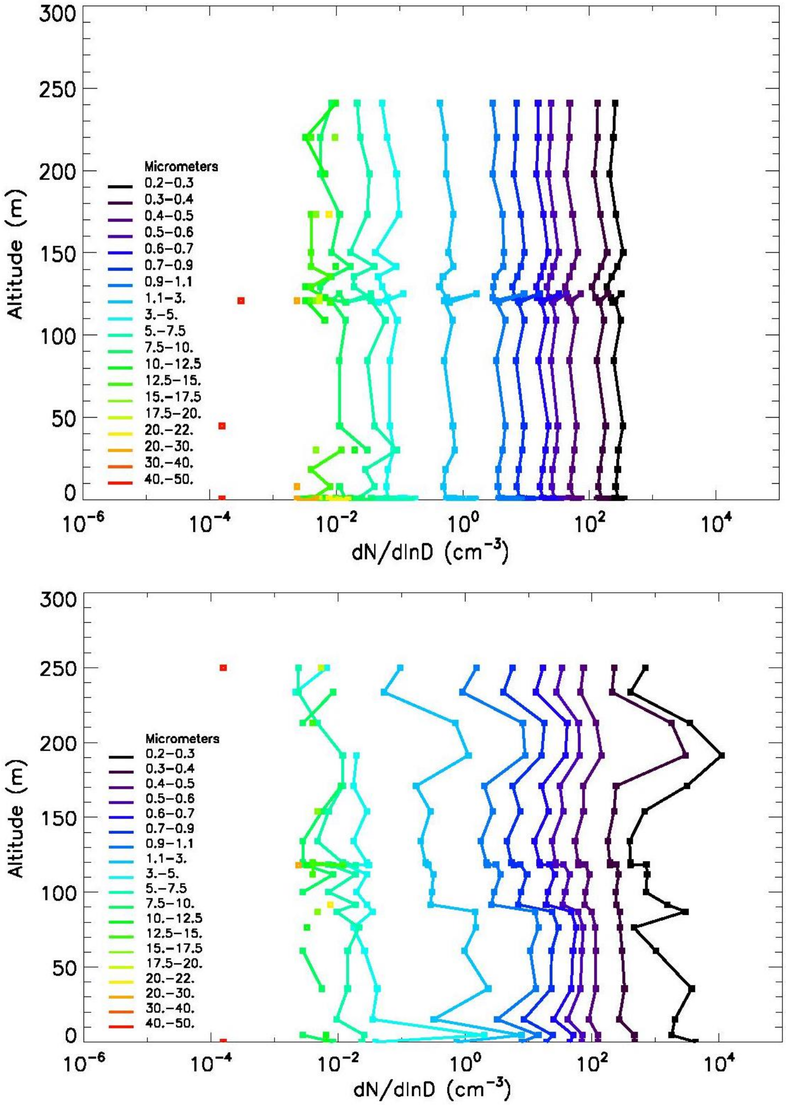

Figure 8: Evolution of the concentrations for the 19 size classes of LOAC, during a flight under the OAG tethered balloon in Paris (France); top: on 10 April 2014 at 9:15 UT (low level of pollution); bottom: on 11 December 2013 at 10:15 UT with a high level of fine particles pollution; the uncertainty (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}

The validation for ground-based measurement at OAG was done in the paper 1 for mass concentrations. On the other hand, there is no possibility to conduct cross-comparison for measurements under balloons since, as for Vienna observations, no measurements were available with altitude for the size distribution.

Unfortunately, the air temperature was not recorded during the flights (the existence of an inversion layer was confirmed by meteorological analysis).

LOAC can detect large particles, as shown in the paper for unmanned aerial vehicle (Figure 3) and tropospheric pressurized balloons (Figure 13) measurements, but no particle greater than 20 μm was detected during the flight presented here.

The TSP inlet remove all particles greater than 100 μm (as said in paper 1); this properties was confirmed during laboratory tests.

L. 2-3 p.10066 and Figure 9: In the upper panel, the part of the "speciation index curve" included in the "Mineral" region includes mainly particles in the 0.2-0.8 μm diameter range. As seen in Paper 1 (cf. my comments on the paper and e.g. comments on Figure 4), LOAC seems very insensitive in this size range, so that the behaviour of the "speciation index curve", which shows an outlier behaviour in almost all illustrated cases, including the ones in Paper 1. Further, as mentioned by the authors in Paper 1, detection of mixed aerosol types using this "speciation index" method is inherently particularly hazardous. Consequently, the interpretation in terms of "mineral aerosols" is very uncertain, what should be mentioned. See also my comments on Figures 18-22 in Paper 1.

The LOAC detection capability in the 0.2-0.8 size range was confirmed by the new laboratory measurements presented in paper 1, during a cross-comparison exercise with a SMPS instrument.

We disagree with the referee comment. The detection of the typology of the particle is not inherently particularly hazardous in case of a mix of mineral and carbon particles. The "speciation index" really indicates a mixture of such particles, the index being located at the boundary of mineral and carbon zones.

3.4 ChArMEx tropospheric flights

Figure 11 and 12: The very different layout and positioning of both figures make them not very clear. The authors should limit the picture in Figure 12 to the same geographic area as in Figure 11, and indicate the balloon trajectory on the figure. Figure 11 can then be removed.

The referee is right. We have removed figure 11 and plotted the trajectory on figure 12 (now called figure 11).

3.5 Upper tropospheric and stratospheric flights

The feature between 5.5 km and 8 km in Figure 15 is unclear: is there any measurement in this altitude range? If yes, why did particles higher than 0.7 μm disappear? And if no, which kind of information was used by the authors to claim that the sand plume was up to 7 km?

Figure 15: There is a lack of measurements between 5.5 and 8 km due telemetry loss. We propose to remove this profile and to plot another tropospheric profile obtained during another flight inside a desert dust plume, on 18 June 2013 afternoon. We plot only the part of the profile where the plume was detected, for clarity reason. We have changed the text to:"

Figure 15 presents the flight conducted from Minorca (Spain) on 18 June 2013 between 16:30 and 17:00 UT (only the tropospheric part of the flight is presented for clarity reason). A mineral dust plume was detected in the lower troposphere up to an altitude of 5 km, with a strong enhancement of aerosol concentrations. The typology analysis confirm the mineral nature of the particles (Figure 15, top), while mainly carbon particles dominate the aerosol content just above the dust plume layer (Figure 15, bottom)."

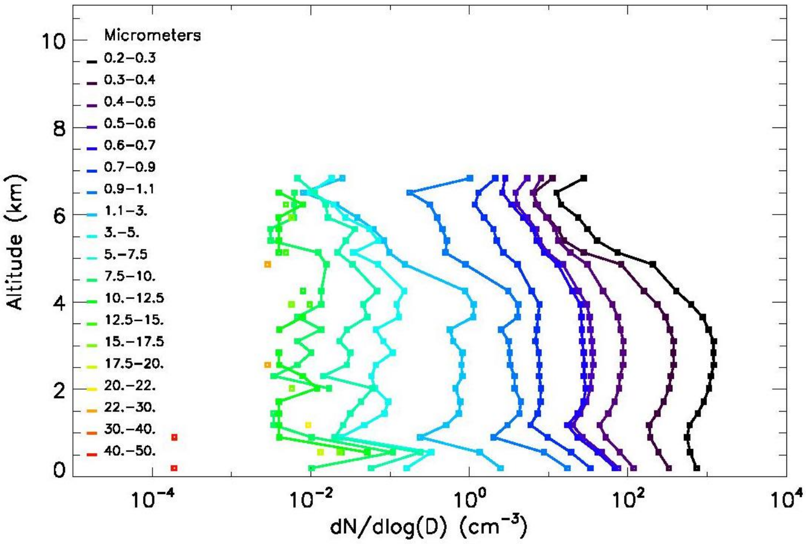

Figure 15: LOAC flight under a meteorological balloon from Minorca (Spain) on 18 June 2013 between 16:30 and 17:00 UT during the ChArMEx campaign; the uncertainty (at 1σ) is of about ± 20% for concentrations higher than 10^{-1} cm^{-3} up to ± 60% for concentrations lower than 10^{-2} cm^{-3}

We have also changed the 2 topology figures to:

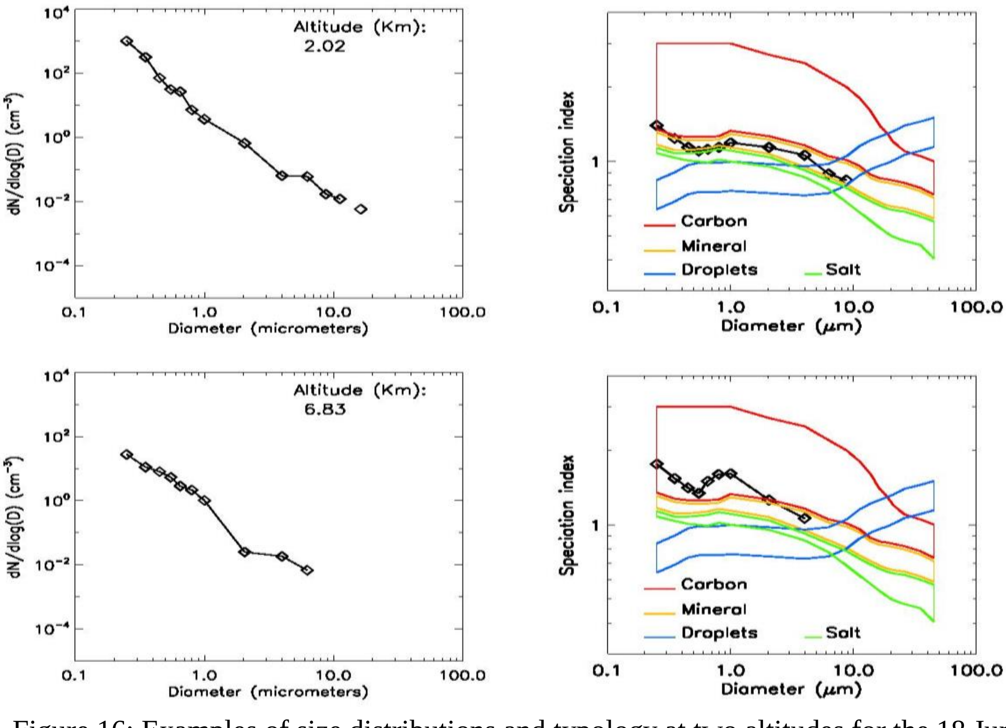

Figure 16: Examples of size distributions and typology at two altitudes for the 18 June 2013 2014 flight from Minorca (Spain); the LOAC data are black dots. At an altitude of ~2 km the typology indicates mineral particles; at ~7 km, the typology indicates carbon particles for the smallest size

L. 10-14 p.10069: This is a kind of « take-and-carry » statement! The cited papers report various different campaigns taking place at totally different times and locations. Hence, "Good agreement" should be replaced at best by something like "plausible following results of stratospheric campaigns performed by…". Looking more closely at the granulometry, LOAC's measurements show in the stratosphere a quite high amount of particles of size ~1 to 5 µm, which is absolutely not found in Deshler's measurements [See Deshler et al., 2003 but also Kovilakam and Deshler, J. Geophys.Res., 2015 about corrected measurements from Wyoming].

We agree that the cited paper reports on various campaigns at different dates and locations and we must be careful concerning our conclusions. Thus, direct comparison on the size distribution cannot be made and we don't see the need to give Kovilakam and Desher (2015) reference. However concentration values measured by LOAC correspond to the expected magnitude for aerosol background conditions in the stratosphere. We have changed the text into: "LOAC measurements reveal concentrations mainly below 1 cm^{-3} for particles greater than 0.2 µm, which corresponds to the expected situation for background aerosols in the stratosphere (Deshler et al., 2003; Renard et al., 2010b)."

L. 21-24 p.10069: The profiles rather show a succession of pronounced 2-3 km thick layers with maxima decreasing slowly with increasing altitude, (about 1 orders of magnitude between the maximum peak situated at ~16 km and the highest altitude around 30 km). As a comparison, vertical profiles of background stratospheric aerosols shown in Figure 3b of Deshler, 2003 (cited by the authors) show a single peak around 11 km, and basically a monotonically decreasing concentration with increasing altitude above this peak. The ratio between concentrations at the peak and ~30 km height is about 4 orders of magnitude and, again, no particle larger than 1 µm is detected above 18 km with a concentration ≥ 10-3 cm-3. In [Renard et al., Appl. Opt 2005, Fig. 6], vertical profiles measured above Aire sur l'Adour give similar concentrations for the various particle classes, but not such a strongly stratified structure through the whole vertical profile. Hence, I am not sure that Figure 17 illustrates a "typical example of background stratospheric aerosols".

The referee is right; this profile seems to contain some small enhancements, as those previously detected by the STAC aerosols counter (Renard et al., 2008; 2010). Thus, we propose to replace the profile by another profile (17 April 2014, from Aire sur l'Adour, France), which exhibits low aerosol content in the stratosphere.

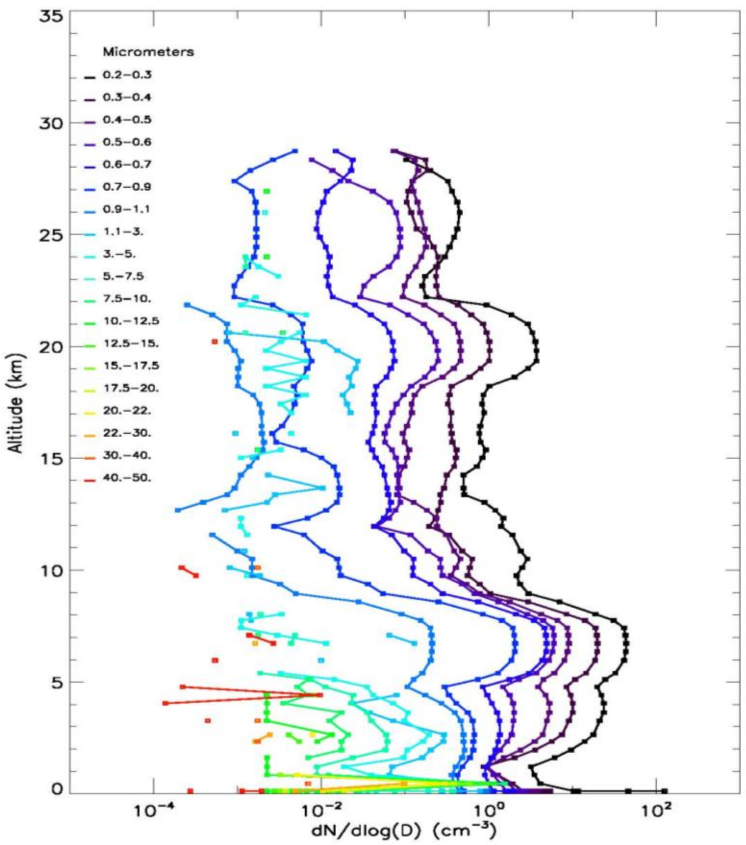

Figure 17: LOAC flight under a meteorological balloon from Aire-sur-l'Adour (France) on 12 August 2015 between 9:50 and 11:40 UT during the VOLTAIRE-LOAC campaign; the uncertainty (at 1σ) is of about ± 20% for concentrations higher than 10^{-1} cm^{-3} up to ± 60% for concentrations lower than 10^{-2} cm^{-3}

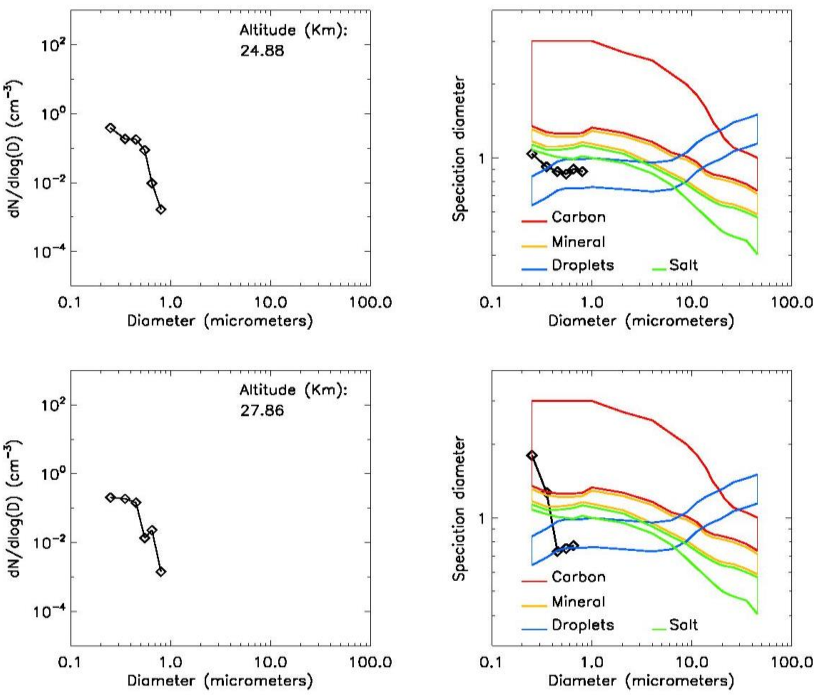

Figure 18: Examples of size distributions and typology at two altitudes for the 12 August 2015 LOAC flight from Aire-sur-l'Adour (France); the LOAC data are black dots. At an altitude of ~25 km, the typology indicates mainly liquid particles; at an altitude of ~28 km, the typology indicates also the presence of carbonaceous particles at the smallest sizes

We have changed the text to "Local transient concentrations enhancements at around an altitude of 30 km were probably observed in the past with the STAC aerosol counter (Renard et al., 2010b), but the nature and origins of particles are not yet fully determined. Such enhancements seems to be also present in some flights performed during the ChArMEx and the VOLTAIRE-LOAC campaigns, but need yet further analysis to be confirmed."

L. 12 p.10070: Again, the authors should be more precise in their quantification. The extinction ratio between values at the aerosol peak and 30 km can be of several orders of magnitude.

The concentrations increases (in the stratosphere) will not produce an extinction ratio increase of several orders of magnitude. Calculations show that they can increase by no more than a factor 30. Note that such increase is not present in the 12 August 2015 profile.

We have then updated the table 1:

Campaign	Launch location	Launch latitude and longitude	Launch date	Balloon type	LOAC inlet
EGU	Vienna (Austria)	48.2343°N 16.4132°E	11 April 2013	Tethered balloon	Metal, bevelled
OAG	Paris (France)	48.8414°N 02.2740°W	11 December 2013	Tethered balloon	TSP
OAG	Paris (France)	48.8414°N 02.2740°W	10 April 2014	Tethered balloon	TSP
ChArMEx	Minorca (Spain)	39.8647°N 04.2539°E	18 June 2013	Tropospheric pressurized balloon	Metal, bevelled
VOLTAIRE-LOAC	Aire-sur-l'Adour (France)	43.702°N 0.262°W	12 August 2015	Meteorological balloon	Metal, bevelled

Table 1: LOAC balloon flights illustrated in this study

Technical corrections:

L. 26 p.10050, L. 20 p.10064: incorrect word.
Correction done.

L. 17-19 p.10061: meaningless sentence.
We disagree, the sentence is correct (but one "," is difficult to read at the end of the line).

L. 20 p.10064, L. 27 p.10065: incorrect units.
Correction done.

L. 8-10 and 10-12 p.10069: sentence revision needed.
Our original text was correct. It is typo errors done by the publisher.

Figure 11 and 12 could be cropped and reduced to the same scale to make the comparison easier and to save place.
We have now merged the two figures.