Answers to Reviewer 2

We want to thank the reviewer for these useful comments that have helped us to significantly improve the paper.

The complex origin and spatial distribution of non-pure sulfate particles (NSPs) in the stratosphere, by Jean-Baptiste Renard et al. gives an overview of the literature on stratospheric aerosol. With more than 100 cited papers a lot of diverse information about stratospheric particles is compiled. In this "review"-part (chapter 1,2,4,5) specific results from diverse papers are mixed together to present an overall picture. But in some cases this presented overall picture is inaccurate as not all presented results can be mixed in reality because of a complete different data base in the cited papers (e.g. particle composition of small carbonaceous particles presented by Ebert et al., 2016 on the one hand and Schütze et al., 2017 on the other hand. They describe completely different particles, which cannot be merged). One conclusion of this manuscript is that data on NSP in the stratosphere is limited and very heterogeneous in dependence of different variables (time, location, height etc.). This is true but this problem cannot be solved by the merge of individual published observations from different locations as presented in this manuscript.

Answer: The reviewer is right; it was the aim of the review of previous works on NSPs to show this problem. We have added at the beginning of part 4: "Although sulfate aerosols are the main component of aerosols in the lower stratosphere, several authors have suggested that NSP could be present in the whole stratosphere. Nevertheless, an overall picture is difficult to establish because results on concentrations, size distributions and compositions stem from different works using different methodologies, and can concern different origins and compositions of particles, which cannot be merged. Thus, the results presented below show the complexity of the NSP content, which is not fully assessed by the studies already conducted."

We have also added at the beginning of part 4.4: "A large variety of shapes and compositions of NSPs have been reported by various authors, and some interpretations on the origins of these particles have been proposed."

Following the cited papers it is obvious how difficult it is to receive evidences (e.g. a link of specific source of NSP to a specific particle size or chemical species). Nevertheless, in this manuscript sometimes the "best guess" is presented as new finding.

Answer: It is difficult to answer. Lots a work needs however to be still done in further studies to confirm our first work. We hope that all the answers we have given to reviewer 1 will satisfy this this comment.

In chapter 3,6 and 7 new data (LOAC) from balloon measurements are presented (sections 3, 6, 7). This data is presented very shortly and it is not really embedded in the review part. It is mentioned that 135 flights were carried out, but in Figure 1-7 the results of only 3 flights are shown.

Answer: Since we present new measurements, these data cannot be embedded in the review part. On the other hand, indeed the LOAC data were presented too briefly.

We have added at the end of section 3.2: "LOAC can be used frequently to performed measurement in the stratosphere to evaluate the vertical and temporal variability of the aerosol content, to identify the presence of NSPs, and also better determine the concentration of large particles greater than 5 mm and up to 50 mm."

We have added in the section 3.3 a figure showing the total concentrations for all the flight and the text: " The mean vertical evolution of number concentrations of particles greater than 0.2 μm can be estimated considering all LOAC flights. At each km, the histogram of the concentrations is calculated and is fitted, in log scale, with a lognormal function to estimate the mode corresponding to the most frequent concentration. As expected for background stratospheric aerosols, the vertical profile of this mode, in red in Figure 2, decreases with increasing altitudes. The individual profiles fluctuate on both sides of the mode profile, probably due to the local variability of the aerosol content. Nevertheless, the variability is higher for the greatest concentrations, with several strong increases of km-width. Such kind of increases reflects those previously reported by Renard et al. (2010) in the middle stratosphere using another balloon-borne aerosol counter.

Examples of individual profiles are presented below, to illustrate the variability of the stratospheric aerosol content."

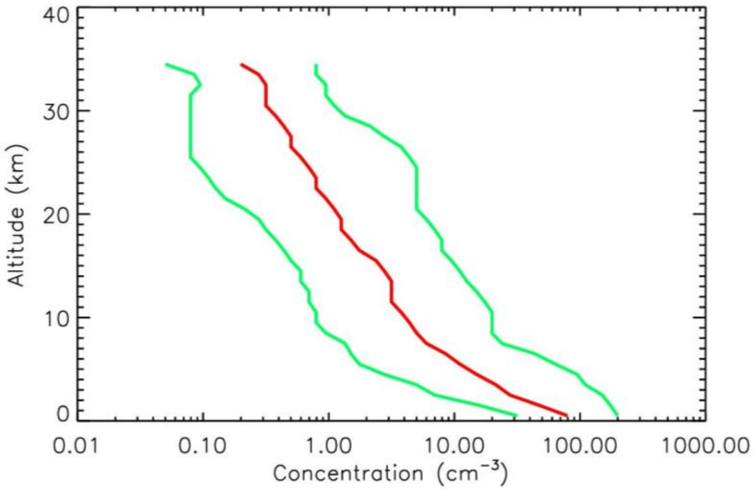

Figure 2: Vertical evolution of the concentrations of particles greater than 0.2 μm considering all LOAC flights; the red line corresponds to the mode of the most frequent concentrations and the green lines correspond to the first and third quartiles

No details about these measurements are given (error sources, artefact discussion, data interpretation), no quantitative data at all (data tables of original data or deduced values)), nor detailed discussion in which way the data is linked to specific questions (beside qualitative speculations).

Answer:

Error sources:

The uncertainties measurements (and thus the errors sources) were already given in part 3.2. We have added: "The uncertainties in the size determination 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."

It is not here the place to discuss in detail the artefact of aerosol counters since it is a well-known technique of measurements. We have already written at the beginning of part 3.2: "Conventional aerosols counters typically performed measurements at large scattering angles (greater than 30° and often around 90°). Since the scattered light at these angles is sensitive to the size of the particles but also to their complex refractive index and their shape including porosity effects (e.g. Muñoz et al., 2001), conventional optical counter measurements must be corrected for the nature of the particles. On the opposite, LOAC performs measurement at small scattering angles, in the 11°-16° range, where the scattered light by irregular particles is mainly coming from diffraction that does not depend on the complex refractive index nor on the porosity of the irregular-shaped particles (Lurton et al. 2014; Renard et al., 2016a)."

If the reviewer is referring to the concentrations enhancements, we have already written: "During the same flight, we consider that a strong concentration enhancement is detected when the concentrations are at least 5 times higher than concentrations above and below the enhancement, for at least 3 consecutive size classes of submicronic particles (this criterion is to ensure that the enhancements are real and are not due to noise measurement fluctuations). We exclude the measurements conducted at the edge of the polar vortex where the local dynamical variability can affect the aerosols content (Renard et al., 2008).”

Data interpretation:

We have tried to improve the discussion on data interpretation. We provide now a figure of the size distribution for the two examples of concentration enhancements.

Indeed, we have not provided tables of the results since we prefer to present figures. Interpretation of the data are presented in Figures 12 to 14.

Concerning the concentration enhancements, we had initially written: “The LOAC typology measurements indicate that the enhancements are dominated by NSPs particles (more than 60%).” And also: “about 25% of the 151 LOAC and 21 STAC flights exhibit strong concentration enhancements”. We have added: “It seems that no obvious correlation can be found between the variability of the number events and the meteor shower dates and intensities, although some fortuitous coincidence can exist. Only 15% of the enhancements occur in a period starting 3 days before the maximum intensity of the meteor shower and ending 7 days later.” To link our results to specific questions, we have added: “Different dynamical processes may be proposed to explain the various concentration enhancements detected by our aerosol counters and previously reported by other authors at different dates and from different locations.”

Concerning the vertical distribution of the enhancements, we had initially written (including deduced values): “A statistical analysis of the altitudes of the concentration enhancement events detected by LOAC seems to indicate a bimodal repartition, one centered at around 17 km and the second one at around 30 km added”. We have added: “This bimodal repartition looks like the one proposed by Beresnev et al. (2017) for the accumulation layers of fractal particles that could correspond to dense aggregates (density of 2 g.cm^{-3}) and of fluffy aggregates (densities in the 0.16-0.35 g.cm^{-3}). In a steady-state atmosphere, under the action of the solar photophoresis the accumulation layers could be in the 13-23 altitude range for the dense aggregates, and in the 25-30 km altitude range for fluffy aggregates, thus close to the ones detected by LOAC and STAC (the LOAC typologies indicate that indeed the optically-absorbing particles dominate the aerosol-enhanced layers).”

Concerning the large particles, we have added: “Finally, LOAC can be tentatively used to estimate the background content of large particles with sizes ranging from several µm up to 50 µm, which are obviously NSPs since large pure droplets cannot exist in the stratosphere.” We have also added: “Since 151 flights were performed in the mid-2013 – 2019 period, it seems statistically reasonable to calculate the mean values of particle concentrations for three broad size classes (5-7.5 µm, 10-17.5 µm, 20-100 µm) in layers of 5 km width. Figure 14 presents the altitude dependence of the mean concentrations; the horizontal bars represent the interannual variability. The concentrations decrease by about of factor two from 15 to 35 km for the three size classes. Ebert et al. (2016) have collected during the RECONCILE campaign inside the polar vortex about 10^3 particles m^{-3} greater than 3 µm in the lower stratosphere (below 21 km). At such altitude, the LOAC concentration of particles greater than 5 µm is below 150 particles m^{-3}; the concentration for particles greater than 3 µm increase to about 400 particles m^{-3} (although some of them could be liquid), which is not so far from the Ebert et al. (2016) value.

Using the Brownlee (1978) measurements, Hunten et al. (1980) have have estimated the concentration of interplanetary dust (or micrometeorites) at 30 km to be of about 10^{-3} particles m^{-3} for sizes greater than 5 µm, On the other hand, the concentration of collected particles greater than 5 µm by the DUSTER instrument during one balloon-flight at 38 km was of about one particle m^{-3} (Della Corte et al., 2013). These concentrations are well below the LOAC mean value of about 50 particles.m^{-3}, although the annual LOAC interannual variability does not indicate any of large particles at such altitude can sometime occur.”

I was looking forward for the data of NSP from 135 balloon flights. But there is no quantitative new data presented in the manuscript. If the manuscript should be more than a pure literature review, more details and discussion to the LOAC data has to be presented and this part must be harmonized with the review part.

Answer: We have tried to significantly improve the presentation and the discussion of the LOAC data, and we give now more quantitative results. The presence of the accumulation layers, and their altitude dependence, the absence of correlation between concentrations enhancements and meteors showers, and the detection of large particles by an aerosol counter and their concentrations are new data. We have also indicated that almost all these particles have more probably a terrestrial origin. We have added at the end of the introduction: "As studying NSPs in the stratosphere is a complex topic, an extensive review of the state-of-the-art techniques is needed. This paper however does not limit to a literature review on this topic."