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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "The complex origin and spatial distribution of non-pure sulfate particles (NSPs) in the stratosphere" *by* Jean-Baptiste Renard et al.

Anonymous Referee #1

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The complex origin and spatial distribution of non-pure sulfate particles (NSPs) in the stratosphere, by Jean-Baptiste Renard et al.

Referee report

General Comments

Starting as a nice overview of the literature on stratospheric absorbing aerosol, this paper becomes much less convincing from the moment that experimental results are analysed, and mainly interpreted. The authors often provide personal interpretations as if they were evidences, without the necessary care. Their references to the literature is also sometimes suffering from a biased or incomplete report on what is written in the

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cited papers. The authors lump together carbonaceous aerosols, black carbon, meteoritic smoke particles, and particle containing non-volatile residues. Further (Section 5.3), they put the emphasis on unusual processes without any reference to their real impact or importance. They give a perception of “huge amounts of absorbing particles” without real distinction amongst them, while there are large differences in optical and microphysical characteristics e.g. between a sulfate aerosol droplet that grew on a non-volatile condensation nucleus (possibly of meteoretic), soot particles from anthropogenic origin, or meteoritic smoke particles. The general view they are giving on stratospheric aerosols is sometimes very chaotic, and in some cases, biased and not in accordance with the literature they are citing. Nowhere, the authors give any perspective about the exact importance of all kinds of absorbing particles with respect to the entire stratospheric aerosol population.

Furthermore, the style of this paper alternates between a review paper (sections 2, 4, 5) and the presentation of personal work around LOAC (sections 3, 6, 7), for which the authors do actually not provide any serious analysis nor any solid, rigourous, and convincing results. LOAC results presented here have a particularly poor information content. The article includes a lot of speculative interpretations without any attempt to validate them using some quantitative data, what is particularly pernicious: the risk is high that conjectures without any solid ground are further cited without too much care (as the authors sometimes do here) and become supposed pieces of truth after a few citations.

The authors should much more valorise this review effort by starting from the categories identifies in Table 1, and to analyse the different aspects (particle size, chemical composition, etc.) for each identified aerosol type. They should clearly separate this review and the LOAC results, and if they choose to use these results, they should propose a solid, grounded, in-depth and quantitative analysis. Anyway, this paper needs a lot more rigour, e.g. through appropriate quantifications of their statements.

Specific comments

L. 61: I suggest slightly modifying the formulation to take into account the influence of the volcanic eruptions mentioned in the preceding sentences, since such eruption (e.g. Nabro in 2011) can really bring temporarily the atmosphere out of this background situation. For instance, the authors could add “out of such volcanic events”.

L. 73: Do the authors mean “pure solid or liquid particles”?

L. 105-108: This sentence is very similar to the sentence on L. 26-29, and in some extend, to the one on L. 1016-1018. The authors should avoid such redundancy. They can explain, for instance, that this section proposes an overview of studies on NSPs detection using all kind of platforms listed in the previous sentence.

L. 133: “(. . .) which backscatter light is well-known”: what do the authors mean?

L. 151-152: This sentence might requires citations.

L. 160-161: This sentence requires also a citation. I guess the sensitivity to the complex refractive index depends on the measurement principle used in in the particle counter. Maybe the text should be qualified in this sense.

L. 162-164: This is actually a very general issue in aerosol retrieval from optical measurements, also by remote sensing.

L. 167-169: “The fraction (. . .) not composed entirely of (. . .) “is a strange formulation. I guess there are other possibilities than water and sulphuric acid to form volatile materials. The authors should thus qualify their sentence (e.g. “i.e. mainly water and sulphuric acid”). “The difference between both channels”: difference of what? Please be more specific.

L. 172: “is used”: I guess the list of measured parameters depend on the study and on the instrument. “Can be” might be more exact.

Section 2.5: Although it is suggested in L. 225-226 through the mention of the assumption on the refractive index, it would be worth to explicitly mention that satellite measurements in the UV-visible spectral range cannot provide information on the aerosol composition (contrarily to satellite measurements in the IR such as MIPAS).

L. 263-266: What do the authors mean? The use of a priori hypotheses on shape, composition and size distribution is necessary. Do the authors mean: “the impact of the use of a priori hypotheses”? Concerning the citation of Bourassa et al., 2012, the authors might usefully prefer Rieger et al., Stratospheric aerosol particle size information in Odin-OSIRIS limb scatter spectra, Atmos. Meas. Techn., 7, 507-522, 2014 where a discussion of the retrieval sensitivity to such parameters is discussed.

L. 268: For the completeness, I suggest that the authors also cite Kovilakam and Deshler, On the accuracy of stratospheric aerosol extinction derived from in situ size distribution measurements and surface area density derived from remote SAGE II and HALOE extinctions measurements, J. Geophys. Res. Atmos., 120, doi:10.002/2015JD023303, 2015. This paper presents an important correction brought to the processing of the optical particle counting measurements by the team of the University of Wyoming.

L. 322-323: The effect of porosity effects is less taken into account in size retrieval studies, and a citation should be required.

L. 328-332: The number and range of provided size classes (19 sizes classes in the 0.2-50 μm) is very large, and it might be useful to give some more insight on the uncertainty as a function of the size range. In addition, I recommend providing a reference where the uncertainty aspects of LOAC measurements are discussed.

L. 344-354: This requires a citation.

L. 369-371: Do the authors mean that a different kind of balloon is used at Aire sur l’Adour, or is it some kind of nomenclature used at this place or by CNES? Please clarify.

L. 375: The profiles in Figure 2 do not show monotonic decreasing profiles with altitude. Please be specific.

Figure 2: Are the points around a concentration range of 1.E-7-1.E-5 cm-3 significant, or are they under the detection limit of the instrument? In the first case, please justify with references to a paper describing the specifications of the instrument. In the latter case, I suggest to remove them from the plot and to adjust the scale for the sake of clarity.

Caption of Figure 3: In my opinion, the authors should give a more detailed description of the figure, and more particularly of the meaning of the coloured regions.

L. 380-383: This explanation seems related to Figure 2. It should thus come before the mention of Figure 3. Interpretation in L. 381-383 is highly speculative and not sufficiently scientifically grounded: I guess many other reasons might explain the presence of one large particle at that place and time. The authors should remove this statement lacking of evidence.

L. 399-403: This criterion is not clear. How do the authors measure the “background” used as reference to assess the “strength of the concentration”. Are they comparing values of the size distribution at a same altitude? This needs to be specified. Do the authors mean that they try to find bimodal size distributions, or discontinuities within a size distribution?

L. 409-411: Again, the interpretations of the origin of these particles are highly uncertain and the authors should just be content with the mention that, in view of the speciation index, these particles are likely to be absorbing, in agreement with Figure 5. Examples of possible composition inferred from the database mentioned in L. 345 (e.g. carbonaceous particles) might be given, but with appropriate references to other works to support their conclusions. As shown here, only the character “transparent”, “semi-absorbing” or absorbing” may be inferred.

L. 410-411: It is hard to distinguish something special between 8 and 12 km: For some particle classes ($\sim< 0.5$ μm) the vertical profile is increasing toward lower altitudes, for other ($\sim$0.5-3 μm), it present a peak at 8 km, and for larger particles (~3-50 μm), it decreases toward lower altitudes with typical concentrations of ~1.E-3-1.E-4 cm-3. Furthermore, these results are very different from results of cirrus size distribution at mid-latitude presented by Zhao et al, J. Am. Met. Soc. https://doi.org.10.1175/2010JAS3354.1, finding typical size distributions with a maximum concentration around 5 to 30 μm from airborne in situ measurements and remote ground-based measurements, while the size distributions found here between 8 and 12 km present a minimum around 10-40 μm. Hence, the authors should present convincing arguments to interpret these particles as cirrus clouds, or remove this sentence.

Figure 7: It is strange that, contrarily to Figure 5 where two clearly different groups of points, spread in different "speciation zones", suggest at first sight the present of two different modes, in Figure 7, all indicated points seem to belong to a same group, spread over the 3 provided zones and even out of them. This seems to suggest, either that LOAC measurements cannot analyse adequately such case, or that another particle type might not be represented on the plot. Also strange is the fact that the yellow zone is obviously different in Figure 5 and 7, while the red and blue zones look similar. What is the reason for this discrepancy?

L. 437-447: "As a result" of what? I really don't know from which element the authors can conclude that LOAC and STAC measurements detected similar concentration enhancements: so far, no single STAC measurement was presented! Concerning the "strong gradients" at the considered altitudes in Figures 4 and 6, there is indeed a sharp structure showing a clear change in the air masses. However, it seems that all vertical profiles roughly undergo a jump of a similar amplitude, possibly suggesting that mainly the total particle number density is changing, but that the shape of the particle size distribution is hardly affected. This would undermine the conclusion drawn in the present discussion. Therefore, it is very important that the authors provide such an illustration of the particle size density as a function of the particle size, for the considered altitude and for the closest altitude levels (say, 3 or 5 altitude levels around 18 km for Figure 4, and around 28 km for Figure 6). In this way, they could verify that the size distribution is clearly different at the level presenting a strong gradient. Also, if the authors want to make some statistics about “strong enhancements” (L. 439-440), they should provide some quantitative criteria to define such observation. They should also provide uncertainty estimates to correctly assess the strength of the phenomenon. Overall, the conclusion that most of the concentration enhancements corresponds to a dominating population of NSPs is clearly premature.

L. 459-462: If many research works (including the ones cites by the authors) show the importance of meteoritic smoke particles, this statement is much too simplistic in a paper on “the complex origin” of NSP. Also, although the mentioned authors indeed study the role and importance of meteoritic smoke particles in the atmosphere (often in a much broader scope than this particular region around 40 km altitude), I did not find in these papers any statement that extinction enhancements at these altitudes are definitely due to meteoritic smoke particles. The authors should thus be much more cautious in their mentions of the cited papers, and they should qualify their statement (e.g. “extinction enhancements are likely associated”, or “the dominating role of meteoritic smoke particles has been shown at these altitude” . . .).

L. 463-465: Detection of absorbing aerosol requires techniques able to sound the complex character of the index of refraction, through polarization effects or scattering properties. This is the case of instruments such as MISR, OMI or PARASOL, but GOMOS does not offer such kind of possibilities. Hence, if it is possible from GOMOS to detect extinction with a spectral behaviour unlikely to be due to sulphate aerosols, it is not possible to attribute unambiguously such signature to the presence of absorbing aerosols particles. Please qualify this statement.

L. 482-484: Although Baumgarder et al. (2004) indeed mentions this result, this statement is biased in absence of the complete information (e.g. Figure 4 in Baumgarder et al (2004)) because it gives wrongly the impression that absorbing particles are fully dominating the particle size distribution. This is not true and for particles larger than 0.3 μm, the non-light absorbing particle are largely dominating everywhere. The authors should provide a complete, non-biased information and specify how the whole size distribution behaves.

L. 485-487: “Similarly”: What is similar? Please reword.

Section 4.2: In this section, the authors are putting all together all kinds of events from fires to meteoritic smoke and even a large-size meteor, all of them seemly assembled under a common characteristic of “spectacular event”. I am not sure there is a real scientific interest to consider such a category “sporadic strong enhancements” without much common characteristics of origin, composition, geolocation, or any other common feature that might interest the reader.

Section 4.3: This section is equally a catch-all of a bit of everything. It becomes rapidly clear how the authors lump all kind of absorbing aerosols together. Starting from the case of “meteorite ablation around 80 km and airplane collected particles”, the discussion of this case continues with “smoke particles”, “interplanetary dust” and “meteoritic debris” and becomes “NSPs” in the next sentence. There is thus an obvious confusion between all these cases, although there is no reason at all that aerosol modes as different as smoke particles, soot particles or other NSPs (maybe sulphate aerosols that grew on some meteoritic condensation nuclei) have similar general size characteristics, since their origin, chemical composition and morphology are completely different. Overall, this section looks like a messy catalogue of numbers of which I am not sure about the benefit.

L. 641: “The detection of metallic spheres”: Please cite the corresponding work.

Section 4.4: Is it right that the two first paragraph (L. 630-650) refer to carbonaceous aerosols of anthropogenic origin, while the third one (L. 660-675) concerns meteoritic particles? And what about the last paragraph (L. 676-682)? The effort of reviewing all these works and providing an insight into the wide range of compositions of these aerosol types is very valuable, but it would be useful to split this overview into aerosol types (e.g. soot from anthropogenic activities, meteoritic particles from extra-terrestrial origin, and possibly particles from unknown origin).

L. 697-702: The interest of this isolated mention of "prograde orbits" is unclear. Further, it is difficult to understand why, just after mentioning the difficulty to categorize aerosol types, the authors seem to look for further sub-categories.

L. 714-715: This statement requires revision: although this question was widely debated in the community, it is likely the volcanic plume of the Nabro (13°N, 41°E, June 2011) reached the upper troposphere due to the eruption and was transported afterward to the stratosphere along pathways provided by the Asian summer monsoon.

L. 716: What do the authors mean by "organic fuel"?

L. 722; Tropopause folds are phenomena occurring in the extra-tropics.

L. 720: Which kind of overshooting convective systems do the authors have in mind? The three provided reference correspond to pyroconvection.

L. 725: "largely determine the composition of the UTLS": is it true that the AMA is the determining factor of the composition of UTLS? It might be necessary to quality this statement in "plays a major role in the composition (. . .)". This statement would require a reference in any case.

L. 734-735: This sentence requires some reference. In particular, the authors should mention important projects specifically dedicated to the study of the ATAL and its composition such as STRATOCLIM (Brunamonti et al, Balloon-borne measurements of temperature, water vapor, ozone and aerosol backscatter on the southern slopes of the Himalayas during StratoClim 2016-2017, Atm. Chem. Phys., 18, 15937-15957, 2018) or BATAL (Vernier et al., BATAL, the balloon measurements campaign of the Asian tropopause aerosol layer, B. Am. Meteor. Soc., May 2018, 955-973, http://doi.org/10.1175/BAMS-D-17-0014.1, 2018).

L. 750-756: How significant are such source in the stratosphere?

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L. 757-768: What is the frequency of re-entry of satellites or rocket disintegration? The probability of observing such event looks particularly low to me, and the interpretation of Ebert's results in the light of Hamdan's work looks highly improbable and speculative to me

Section 5.3: It is a little bit strange that the overview of the NSP production mechanisms describes as second mechanism the formation of "hypothetical long-lived volcanic soot particles" and as third one "rockets exhaust and the disintegration of satellites" of which the relative importance is particularly uncertain in the global NSP production. This one is followed by "compact particles and filaments" possibly (hypothetically?) occurring during atmospheric entries of meteorites and satellites/rocket debris" , and "rare events of the disintegration of large meteoroids, with sizes above 10 m". Finally, the very last one mentions meteoritic ablation likely to be about the most important of all the cited processes. The authors should present the different mechanisms in the order of importance and impact on the atmospheric processes, starting from the most important. They also should quantify in some way their importance, in terms of concentration, spatial coverage, lifetime, occurrence rate, etc.

L. 795-797: Sentence is not clear.

L. 797: Interpretation may be arbitrary. Do the author mean "analysis"?

L. 804: "faint dust bands attributed to collisions within the asteroid belt": It is not clear if the authors refer to some particle "cloud", or to some spectral band. Please clarify.

L. 808-810: Odd sentence. Please reword.

L. 812-813: Which cloud? This sentence requires some reference. Idem for L. 813-817.

L. 826-828: Incorrect sentence: What is "prevalence influx"? Speed is a physical parameter, thus singular.

L. 829: "relative velocity": with respect to what?

L. 829-830: The authors misuse results published by Levasseur-Regourd and Lasue (2011). These authors draw their conclusions about the probable "survival" of significant amount of organics for seeding life on early Earth, during the Late Heavy Bombardment epoch, "while the spatial density of dust in the interplanetary dust clouds was orders of magnitude greater than nowadays".

L. 841-842: The meaning of the use of italic characters is unclear.

Section 5.3: The authors give a lot of details about interplanetary dust particles and discuss extensively the origin of these particles (comets or asteroids). However, the conclusions of interest for the study of NSP is particularly poor and vague: enhancements of interplanetary aerosol particles [of carbonaceous aerosols] might show similar properties (which ones ?) as aerosol particles of terrestrial origin. The authors should shorten this section.

L. 866-867, Figure 9: Does it mean that STAC and LOAC flights span several weeks? This is not what previous text suggests (e.g.:, Figures 1 to 7, corresponding to flight on a single day). The authors should clarify what they mean on Figure 9, and how many flights are concerned. A table with an overview of all types of flights and their characteristics might be useful.

L. 863-865: On which criteria do the authors base their statement? How fortuitous such event is depends on its persistence and frequency. Based on the potential total daily amount (up to 270 tons) given in L. 833, meteoritic smoke particles might be not so fortuitous.

L. 869-872: Did the authors calculate any correlation?

L. 871-872: I do not see any convincing, quantitative analysis that might lead to this conclusion.

L. 880-917: The authors list a full page of speculative explanations, without proposing any serious analysis on how their measurements may support any of them. I have some doubt about the relevancy of such approach.

L. 918: “double repartition” is incorrect.

L. 921: “This double repartition looks like the one proposed by Beresnev et al. (2017)”: this is again a very weak “analysis” referring to a non peer-reviewed proceeding, with a vague description (“the [double repartition] for the accumulation layers of fractal and spherical carbonaceous particles respectively”). Such a superficial analysis is totally insufficient.

L. 925-927: What does it mean? What do the authors want to prove?

L. 945-946: Following Figure 11 and taking into account the error bars, there is no statistically significant decrease of the concentration with increasing altitude.

Caption Figure 11: “Evolution” (should be singular) seems more a time-related concept. “Altitude dependence” seems more adequate. What is “mean evolution”? Please specify what the circles represent.

L. 948-949: Even if the ensemble of the considered measurements are likely to provide much useful information, it is impossible to conclude on any trend from the results given in Figure 11 (profiles and their error bars) and the fact that the total analysed volume of air represents about 1 to 2 cm3 of the whole stratosphere.

L. 949-951: What do the authors intend to prove by this statement? What is the available measurement sample and the representativeness of these observations?

L. 952-961: This “analysis” is inadequate. How do LOAC flights match with the RECONCILE campaigns inside the polar vortex? Even by summing the contributions to all panels of Figure 11, the result does not reflect in any way a concentration of about 1000 particles/m3 for particles greater than 3 microns below 21 km. And saying that LOAC measurements agrees with cited results at 30 km and 38 km respectively, is meaningless. The authors should remove all this discussion.

L. 973-975: This reference is difficult to find, and the mentioned rate of 10% is not mentioned in a summary found on internet. However, the title of the paper specifies that this work concerns micrometeorites collected at ground level in ultra-clean snow in Antarctica. This is far from representative of the interplanetary dust flux reaching the (global) stratosphere. The rest of the section (L.975-1011) is an additional hope of speculation without any attempt to test them against any quantitative result.

L. 1030-1034: The developments as presented in the paper do not allow drawing these conclusions.

L. 1072-1076: These sentences include generalities without any relevant added information. I suggest removing them.

Technical comments

L. 27: missing comma.

L. 35-36: “ranging between 17 and 30 km altitude”?

L. 227: “They cannot access the local variability” or “They give no access to the local variability”.

L. 114: “sparse of time-series measurements”: looks odd. Maybe “sparse or continuous”, “sparse or routine measurements”?

L. 118: “abundance”. Physical parameter, thus singular.

L. 127: “EARLINET”.

L. 241-243: Odd sentence (combination of “often” and “in general”).

L. 244-246: “such as volcanic eruptions, injection (. . .), or pyroconvection”?

L. 264: “hypotheses”.

L. 313: “can be”?

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L. 333: I suggest “The particle size provided by LOAC” to avoid the strange association “particles provided by LOAC”.

L. 355: “corrected for”.

L. 373-374: “an example of vertical (. . .)”; “on 17 August 2017”

L. 444, Figure 3 (and maybe elsewhere): “absorbing” instead of “absorbent”.

L. 704: “main characteristics”.

L. 752: missing “)”.

L. 793: Please correct the sentence.

L. 795: “with’ instead of “within”.

L. 817: missing ‘.’

L. 971: “originating”.

L. 1175: “boundary”.

L. 1263: “Fadnavis, S.”.

L. 1330: “Process.”

L. 1595: “OSIRIS”.

L. 1014: “Conclusions” is Section 8.

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