ACP-2020-660
Aircraft-based observation of meteoric material in lower stratospheric aerosol particles between 15 and 68°N
J. Schneider et al.

Reply to Reviewer #2

The reviewer comments are written in this font style and color.

Our answers are written in this font style and color.

Changes to the revised version of the manuscript are printed in red.

Review of manuscript by Johannes Schneider et al. for Atmos. Chem. Phys. "Aircraft-based observation of meteoric material in lower stratospheric aerosol particles between 15 and 68N"

This manuscript presents an analysis of high-altitude aircraft measurements of stratospheric aerosol particle composition from two different laser-ablation aerosol mass spectrometer instruments.

The study brings together aerosol composition measurements from 5 different field campaigns between 2014 and 2018, from three different research aircraft (the German G550 HALO, the European M55 Geophysica and the US NASA DC-8).

The analysis focuses on assessing the prevalence of the iron and magnesium particle spectra within the mid-latitude stratospheric aerosol layer, the composition signature indicating the presence of meteoric material within these particles. This topic is of particular interest given the similar PALMS laser-ablation aerosol composition measurements on the WB-57 high-altitude aircraft showed in 1998 this meteoric signature was highly abundant among aerosol particles in the mid-latitude stratosphere. Refractory particle counter measurements in the Arctic stratosphere show elevated concentrations within the polar vortex, increasing with altitude into the stratosphere, thereby also strongly indicating them likely being meteoric particles transported down from the upper atmosphere.

The authors are to be congratulated for bringing together this breadth of measurements from the different campaigns, which then enables to assess the meridional extent of the fraction of particles with the meteoric composition-signature, and the vertical profile of the abundance of these particles within the lowermost stratosphere. Measurements are shown across the range of latitudes and altitudes sampled in 5 different aircraft missions that sampled in the upper troposphere and lower stratosphere (UTLS).

There has been a renewed debate in the stratospheric aerosol community about the presence of meteoric material since the PALMS measurements in 1998 revealed ~50% of particles in the mid-latitude lowermost stratosphere contained signatures of meteoric material. The refractory particle measurements in the Arctic stratosphere from 2002/3 (Curtius et al., 2005) have been confirmed with similar enhancement in refractory particles within the polar vortex also found in Arctic campaigns in 2009/10 and 2010/11 (see Weigel et al., 2014), consistent with the particles being transported each winter within subsiding air masses in the polar vortex,

bringing a seasonal source of meteoric particles down into the stratosphere each winter.

The observations will also be of considerable interest within the interactive stratospheric aerosol modelling community, with the majority of current models tending only to simulate the homogeneously nucleated particle population, most not including the particle formation pathway of heterogeneous sulfuric particle formation on meteoric particles.

I should disclose that I reviewed an earlier version of this manuscript submitted to JGR in Jan/Feb 2020, a large number of corrections and revisions required at that time, myself and the other two reviewers each finding independently substantial changes were required before publication. The authors replied to all of the reviewer comments, and a greatly improved revised manuscript was submitted, but although I recommended publication after minor revisions, I can only assume the consensus among the reviewers and editor was that the manuscript was still not quite at the standard for publication, as I was subsequently notified that the paper had been rejected.

I can see that the paper has been substantially further improved since that time, in particular with there now being a very welcome additional Supplementary Material section, which presents additional background information to enable the interpetations of the data given in the main article to be further scrutinised and better understood.

The article is certainly suitable for ACP and is generally well-written, and I recommend publication after a number of specific minor revisions are made, which I have listed below.

We thank the reviewer for this generally positive rating of our manuscript.

However, there is one major concern that the authors need to explain, and caveat the "percentage meteoric" values presented in the Abstract sufficiently such that readers understand the real values may be substantially less than this, because of undersampling of the pure sulfuric particles.

We have addressed this major concern in the revised version and explain this in the answers given below.

The sentence beginning on line 483 states "...pure sulfuric acid particles are not ablated and ionized by a laser with wavelength of 266nm, because sulfuric acid has a very low absorption cross-section for wavelengths larger than about 190nm up to visible light", then citing the articles by Thomson et al. (1997), Burkholder et al. (2000) and Murphy et al. (2007). The PALMS instrument used in Murphy et al. (1998) and Murphy et al. (2007) papers uses a 193nm laser, whereas, as is explained in the article, both the ALABAMA and ERICA instruments use 266nm lasers.
The Thomson et al. (1997) article assesses and discusses the aerosol absorption from 157nm, 193nm and 248nm lasers, and reading that paper, I see indeed the issue the authors are referring to --- that there is a large difference in absorption behaviour between pure sulfuric particles and those with organics and other "small amounts of contamination".
The authors state in the paragraph on lines 481-487 that "the fractions of particles containing meteoric material will be overestimated if pure sulfuric acid aerosol particles existed in the air".
But it is far from clear how the reader should then interpret the results.

Are the authors arguing that there are actually very few particles in the lower stratosphere that have levels of impurity low enough for them not to be ablated by the 266nm laser?
If so then they need to replace that wording "if pure sulfuric aerosol particles existed" and state it in those terms.

We rewrote Section 3.5 as detailed below.

As the text stands, this potential undersampling issue remains a potential problem, potentially rendering the proportions stated in the Abstract to potentially be substantial overestimates.

The central question is to what extent particles are "pure enough" to suffer from the effect discussed in the Thomson et al. (1997) paper. Perhaps the authors are explaining that, in reality, all particles in the stratosphere contain a sufficient level of impurity that they believe that there is not a significant undersampling problem.
I do not know the answer to that question.
Figure 3b in the Murphy et al. (2007) shows the mass spectra of the "pure sulfuric particles" (as named in the Murphy et al., 2014 article) measured by the PALMS instrument (with the 193nm laser). Those spectra without the Fe and Mg signature do have other impurities, and it is not clear to me whether or not these would be detected by the ALABAMA and ERICA laser-ablation mass spectrometer instruments.
Provided the authors can provide information to assure the reader that this is the case, or can provide sufficient caveat to explain the severity of the potential undersampling they are acknowledging may be an issue, then the article can be published accordingly.
But the article is currently far from clear about this.

The PALMS instrument, using an ablation laser with 193 nm, is able to detect "pure sulfuric particles", as presented in Murphy et al. (2007; 2014). This particle type reaches a fraction between 10 and 20% at mid-latitudes, for ozone mixing ratios up to 1800 ppb (see Fig. 3a in Murphy et al., 2014). In Murphy et al. (2007; Fig 4), the reported fractions range between 10 and 30%, up to 8 km above the tropopause, with ozone reaching up to 1200 ppb. These campaigns were conducted out of Houston, Texas, and San Jose, Costa Rica.

We may therefore assume that in our data set (ozone never exceeding 1200 ppb), the underestimation of the total analyzed particle number due to the presence of pure sulfuric particles is about 20 (± 10) %.
This translates into an overestimation of the fraction of meteoric particles by the same range.

We state this underestimation in the abstract of the revised version and rewrote Section 3.5 (see below). However, we prefer not to correct for this underestimation, as the uncertainty would be very high. The variation of the pure sulfuric fraction with altitude, potential temperature, latitude, and season is not known well enough to transfer the data by Murphy et al. (2007; 2014) to our data set.

A related point, is that the authors really need to give some information about the composition of the particles ablated by the laser that do not have the Fe and Mg composition signature.
Do the ALABAMA and ERICA measure, similarly to Murphy et al. (2007), that the particles without the Fe and Mg signature group into spectra that are rich in carbonaceous material (i.e. Figure 3c in Murphy et al., 2007) and those that don't (those in Figure 3b)?

Our results give a more complicated picture of stratospheric aerosol. The particles do not only fall into the three categories that were described by Murphy et al. 2007 and 2014, but contain also ammonia, black carbon, secondary organics, and nitrate. We present an averaged mass spectrum of all stratospheric particles that do not contain the meteoric signatures (Mg and Fe) in the new panels (b) and (d) of Figure 2.
However, a further analysis of the different types of stratospheric particles and their composition and sources is outside the scope of this paper and will be presented in upcoming publications.

Murphy et al. (2007) show that the vast majority of the particles within the meteoric signature are the organic-rich sulfuric particles.
In which case the undersampling problem is not so important, because only 10-20% of the particles are pure enough to be missed by the ALABAMA and ERICA instruments.
And those percentages with the meteoric composition-signature can be considered to be highly reliable (albeit with a slight overestimate of perhaps 10% or so).
Basically the paragraph on lines 481-487 needs to be re-written to explain much more clearly how the reader should then interpret the fraction of meteoric particles being presented.

We rewrote the beginning of Section 3.5 as follows:

"It is difficult to estimate accurately the absolute number concentration of particles containing meteoric material from the measured particle fraction with our laser ablation mass spectrometers. The main reason is that pure sulfuric acid particles are not ablated and ionized by a laser with a wavelength of 266 nm, because sulfuric acid has a very low absorption cross section for wavelengths larger than about 190 nm up to visible light (Thomson et al., 1997; Burkholder et al., 2000; Murphy, 2007). Thus, the fraction of particles containing meteoric material will be overestimated due to the presence of pure sulfuric acid aerosol particles in the stratosphere.
The PALMS instrument, using an ablation laser with 193 nm (Murphy et al., 1998; Cziczo et al., 2001; Murphy et al., 2007; Murphy et al., 2014) is able to detect pure sulfuric acid particles. The results presented in Murphy et al. (2007) show that the number fraction of the sulfuric particle type ranges between 10 and 30% up to 8 km above the tropopause and at ozone mixing ratios up to 1200 ppb. These data were obtained at tropical (Costa Rica) and mid latitudes (Texas). In Murphy et al. (2014), the presented number fraction of sulfuric particles measured at mid latitudes ranges between 10 and 20 %, for ozone mixing ratios up to 1800 ppb. We may therefore assume that in our data, where ozone never exceeded 1200 ppb, the underestimation of the total analyzed particle number due to the presence of pure sulfuric acid particles is about 20 %, ranging between 10 and 30 %. This translates into an overestimation of the meteoric particle fraction by the same percentage. However, the variation of the pure sulfuric acid fraction with altitude, potential temperature, latitude, and season is not known well enough to apply a correction to our data set. Thus, it must be noted that the meteoric particle number fraction as well as the following estimation of the absolute number concentration of particles containing meteoric material may be overestimated by 10 – 30 %."

And I recommend adding to Figure 2 (or to the Supplementary Material) equivalent panels showing this mean spectra for the particles without the meteoric signature, with (if possible) further separated into those with and without the carbonaceous signature, consistently with the categorization from Murphy et al. (2014) as "pure sulfuric" and "organic-sulfuric" particles.

We added to Figure 2 the mean spectra of all stratospheric particles without the meteoric signature. A further separation into different particles types (such as with and without carbonaceous signature) will be subject of upcoming publications that focus on the composition of UTLS aerosol under the influence of Asian and African monsoon.

It sounds like the authors are explaining that only a small proportion of the non-meteoric-signature particles are pure sulfuric particles. And that is consistent with the results from the PALMS measurements. If so then I advise that be communicated within a revised version of this manuscript.

We rewrote section 3.5 (see above)

The paper needs to provide the reader with a clear interpretation of the findings, so that an approximate %-confidence measure can be considered in relation to the 20-40% and 60-80% meteoric particle fractions/proportions that are presented in the Abstract.

The current wording of that paragraph suggests a more significant under-sampling of the pure sulfuric particles, compared to the meteoric-signature particles, and this central issue needs to be presented much more clearly to enable the results to be properly interpreted.

We rewrote Section 3.5 (see above), removed Figure S8 and modified Figure 7 accordingly. We mention an overestimation of the meteoric particle fraction by about 10 – 30% in the abstract and in Section 3.5.

Since the results are presented in the Abstract without caveat, I am assuming that the bias is of only a small magnitude (which is what the PALMS measurements suggest). If that is the case then a sentence should be added both to the conclusions and the Abstract to provide clarity on the reason why the reader can be confident that is the case.

We added "It must be noted that the relative abundance of such meteoric particles may be overestimated by about 10 to 30% due to the presence of pure sulfuric acid particles in the stratosphere which are not detected by the instruments used here."
Furthermore, we added "observed fraction" or "of the observed particles" to all places where percentage values are mentioned in the abstract.

Although the article is mostly very well written, by contrast the Abstract seemed much less well-written and requires improvement. In almost all cases however, the revisions are minor wording improvements, but are important to better communicate the study's findings.

The rest of the article is well-written, although the Supplementary Material I found also needed quite a substantial number of minor revisions. The list of minor specific revisions are mostly then for the Abstract and Supplementary Material, and aside from the major revision explained above, the majority of the manuscript is in excellent shape already (perhaps reflecting its improvement after the previous set of reviews in the other journal).

Overall, provided the authors can address this one major issue, better communicating the magnitude of the uncertainty via a corresponding sentence added to the Abstract and Conclusions, then I am happy to recommend publication to ACP once the minor revisions listed below are addressed.

Minor specific revisions
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1) Abstract, lines 20-22 -- This first sentence is a little clunky to read, and the scientific aim of the analysis in the paper is better to be communicated earlier in the sentence. Suggest to move "to assess the meridional extent of particles containing meteoric material" to be immediately after "between 2014 and 2018", then replacing "sampling" with the word "in". Also, suggest to delete "In this paper", beginning instead as "We analyse ..." and delete "conducted" I.e. have the sentence be "We analyze aerosol particle composition measurements from five research missions between 2014 and 2018 to assess the meridional extent of particles containing meteoric material in the upper troposphere and lower stratosphere (UTLS)".

Changed as suggested

2) Abstract, lines 22-24 -- stating "confirm the existence of" is not really appropriate. I know what you mean, but it's more to assess whether the meteoric signature is also present in the lower troposphere. "Confirming the existence of" suggests there is some doubt about whether these particles exist at all, which is not the case. Suggest to delete "are used to" and replace "confirm the existence of meteoric material in" with "show that meteoric material is also present within". Also, the Jungfraujoch site is not sampling lower tropospheric particles, but mid-tropospheric particles, so insert "middle and" before "the lower tropospheric particles", adding also the clarifying additional words ", but within only a very small proportion of particles." Also, the wording of the first half of this sentence needs to be improved. Firstly, the word "datasets" is too general a term, better to say "measurements" and delete "Additional" -- and the phrase "a ground based study" should communicate the location, such as "a mountain-top site" or better still "the Jungfraujoch mountain-top site". Also it makes the sentence easier to read to hyphenate "low altitude" to "low-altitude", with the "from" prior to that word also able to be deleted for better wording. So I mean that I am suggesting that the sentence be re-worded to something like: "Measurements from the Jungraujoch mountain-top site and a low-altitude aircraft mission show that meteoric material is also present within the lower and mid tropospheric aerosol, but within only a very small proportion of particles."

Changed as suggested

3) Abstract, lines 24-25 -- Again suggest slight improvement to the wording here to better link to the previous sentences and make it clear these are the main observational datasets in both the UTLS field campaigns, and from the Jungfraujoch and lower-altitude aircraft flights. This can be achieved by changing the start of the sentence from "Single particle laser ablation..." instead to "For both the UTLS campaigns and the lower/mid-troposphere observations, the measurements were with single particle laser ablation...". Also suggest to change "techniques" to "mass spectrometers" to be more precise, and "were used to measure" to "which enabled to measure". Please also replace "size range" with "diameter

range" or add "diameter" at the end of the sentence, so it's clear those values are diameter values.

Changed as suggested

4) Abstract, line 27 -- Delete the words "particles" (after "147,338") and "measured" (before "in the stratosphere"), better not to state that again, it's implicit from earlier in the sentence and easier to read without these words.

Changed as suggested

5) Abstract, lines 27-30 -- Insert "total" after "Of these", delete "and rare iron oxide compounds", (the mass spectra are detecting the ions, and the same could be said of magnesium oxide, but doesn't need to be), also replacing "together with sulfuric acid" with "together with sulfuric ions". I strongly suggest also to merge the subsequent sentence into this sentence, shortening the 2nd sentence so be a 2nd half of this sentence, i.e. replace ". This particle type was found almost exclusively in the stratosphere (48,610 particles) and is" with ", the vast majority (48,610) in the stratosphere,", also delete the "stratospheric" before "sulfuric acid" at the end of the sentence, also deleting the last word "particles". So I mean I'm suggested to re-word to: "Of these total particles, 50,688 were characterized by high abundances of magnesium and iron, together with sulfuric ions, the vast majority (46,610) in the stratosphere, and are interpreted as meteoric material immersed or dissolved within sulfuric acid."

Changed as suggested

6) Abstract, lines 30-32 -- suggest to replace "particle type" with either "meteoric-sulfuric type" or "meteoric particle type" (or similar). Suggest to again join up the subsequent sentence, and shorten, also providing specific values for the two tropospheric locations -- i.e. replace ". However, small fractional abundances were observed below 3000m a.s.l. in the ..." with something like ", with 0.2-1 \% abundance at Jungfraujoch, and smaller abundances (0.0x-0.0y \%) from the lower altitude Canadian Arctic aircraft measurements.".

Changed to "Below the tropopause, the observed fraction of the meteoric particle type decreases sharply with 0.2 – 1 % abundance at Jungfraujoch, and even smaller abundances (0.025 – 0.05 %) observed during the lower altitude Canadian Arctic aircraft measurements."

7) Abstract, line 32 -- this sentence is strange -- it is not a new result to confirm that the removal pathway is by sedimentation and/or mixing into the troposphere. The fact that there is a steep gradient across the tropopause confirms that the particles originate from the stratosphere or above, but that is not the way this is reported. It's kind-of obvious that a tracer with a source in the stratosphere (or above) would have a gradient across the tropopause, and that it would be removed by mixing into the troposphere. The question is really how important sedimentation is, in additional to simply air mass exchange from the stratosphere into the troposphere -- but that's not really addressed directly here. The size distribution of the meteoric-signature particle is however an indirect measure of how important sedimentation is, because if the signature were found only in the smallest particles (~200nm) then

sedimentation might not be that important, but here the findings from Murphy et al. (2014) are confirmed, that the meteoric-signature is found mostly in sulfuric particles at around 400-500nm, with much fewer in the 200-400nm size range. That does suggest that the sedimentation is important in addition to mixing of air into the troposphere. The size distribution of the meteoric signature is not currently mentioned in the Abstract, and this sentence is where this could be stated. I suggest the authors replace this sentence with "The size distribution of the meteoric-sulfuric particles measured in the UTLS campaigns is consistent with that measured by the PALMS measurements, with only 5-10% fractions in the smallest particles detected (200-300nm diameter), but with substantial (> 40%) abundance-fractions for particles from 300-350nm up to 900nm in diameter, suggesting sedimentation is the primary loss mechanism." Or similar sentence to this.

Changed as suggested, but as we did not mention ALABAMA and ERICA up to now, we also would not like to mention PALMS, and instead wrote "with earlier aircraft-based mass spectrometric measurements,..."

8) Abstract, line 36 -- replace "present in higher" with "present at much higher".

Changed

9) Abstract, lines 38-40 -- I'm not sure this sentence is necessarily the case. In the Introduction (lines 81-84), the authors discuss how meteoric fragments may sediment directly into the stratosphere. In contrast, the sentene here suggests the particles are transported down into the mesosphere only at high latitudes. That predominantly-transport-driven seasonal source of meteoric material is the case for meteoric smoke particles (which tend to only be a few tens of nm), but if there is also a significant source of meteoric fragment particles (in addition to the smoke particles), then there may well be a source at other latitudes too. Indeed the finding on line 36 of the Abstract, that similar abundance-fraction is seen across all latitudes and seasons measured suggests the fragments are a substantial proportion of the meteoric material in the stratosphere. Suggest to move "This finding suggests that" to be the start of the final sentence, and have this penultimate sentence explaining this winter polar vortex mechanism is the case for meteoric smoke particles. With then the sentence after explaining that the findings here suggest that there is another source of particles, in addition to the meteoric smoke. I mean change the start of the the sentence beginning on line 38 from "This finding suggests that the meteoric material is transported..." to instead say "Meteoric smoke particles are transported...", change "is efficiently distributed towards" with "is subsequently transported towards..." and I think the authors must mean "below 440K potential temperature" not "above 440K potential temperature", because that transport tends mostly to occur in the lower part of the polar vortex.

We agree that meteoric fragments are as likely as MSP to explain our observations. But, as we can't distinguish between the two, we changed this part mostly as suggested, but with the last sentence: "By contrast, the findings from the UTLS measurements show that meteoric material is found in stratospheric aerosol particles at all latitudes and seasons, which suggests that either isentropic mixing is effective also above 440 K or that meteoric fragments may be the source of a substantial proportion of the observed meteoric material." The uniform occurrence of meteoric material above 440 K is a strong argument for an effective isentropic mixing also above 440 K.

10) Abstract lines 40-41 -- As per comment 9), I'm suggesting to begin this final sentence "By contrast, the findings from the UTLS measurements show meteoric material is found in stratospheric aerosol particles at all latitudes and seasons, which suggests meteoric fragments may nucleate a substantial proportion of the observed meteoric-sulfuric particles." Or something like this.

Changed to "By contrast, the findings from the UTLS measurements show that meteoric material is found in stratospheric aerosol particles at all latitudes and seasons, which suggests that meteoric fragments may be the source of a substantial proportion of the observed meteoric material."

11) Introduction, line 62 -- replace "in the Earth's atmosphere" with "into the Earth's atmosphere"

Done

12) Introduction, lines 68 and 69 -- Although MSP is almost always used with the third letters' corresponding water in the plural (Particles), it makes it much easier to read to communicate the plural including the lower-case s -- as MSPs. This is similar to way polar stratospheric clouds are referred to as PSCs. So replace the instances of "MSP" on lines 68 and 69 instead with "MSPs".
Also on lines 95 and 97.

Done (throughout the manuscript)

13) Introduction, line 82 -- Similarly you likely have "MF" here as an abbreviation for the plural term "Meteoric Fragments" but again, it's better to say "MFs", in the same way as MSPs and PSCs. Please change "MF" to "MFs" here and on line 84. Also on line 97.

Done

14) Introduction, line 83 -- I'm not sure why you are questioning whether meteoric fragments form here. The preceding sentence begins "As has recently been shown...", so either that sentence needs to be changed to "have suggested" or else this sentence needs to be re-worded. However, the existence of meteoric fragments has been clear since rocket-borne measurements in the early 1960s (e.g. Hemenway and Soberman, 1962), with the fragments terminology having been introduced in the 1950s (e.g. Jacchia, 1955). Suggest to re-word the start of the preceding sentence to "As was hypothesised in the 1950s (e.g. Jacchia, 1955) and shown in measurements from the 1960s (e.g. Hemenway and Soberman, 1962), recently also further established by Subasinghe et al. (2016)...". Maybe it's just to change "were formed" to "are formed" and add "at sufficient particle concentrations" afterwards.

Changed as suggested

15) Introduction, lines 90-94 -- Again, although the term IDP is being used here as the plural term, it's easier to read this making the plural clear as "IDPs". Please change "IDP" to "IDPs" in all instances here, except on line 95 when the term is used in the singular.

Changed as suggested

16) Introduction, line 101 -- Change "Later, aircraft based" to "More recently," or "Much more recently,"

Changed to "more recently"

17) Introduction, line 116 -- Delete "summer" from the "Tropics/sub-tropics" because this seasonal variation is not relevant here.

Done

18) Measurements and Methods, line 124 -- replace "includes" with "analyses", insert "lower" before "stratospheric" and provide a more descriptive word than "data", also avoiding using bland terms such as "obtained" (since they don't communicate these being measurements from the field). Suggest also to replace "data obtained during" with "aerosol composition measurements taken" and replace "research" with "field". Also insert "additional composition measurement" after "with two" and insert "the lower troposphere" before "altitudes below 3600m a.s.l.", putting that last text in brackets -- i.e. "the lower troposphere (altitudes below 3600m a.s.l.".

The sentence reads now: "This study analyses lower stratospheric and upper tropospheric aerosol composition measurements taken during five aircraft-based field campaigns, together with two additional composition measurements from the middle and lower troposphere (altitudes below 3600 m a.s.l)."

19) Section 2.1.1, line 133 -- suggest to insert ", the full dataset from" before "which are included".

Done

20) Section 2.1.3, line 147 -- hypenate "aircraft chasing" to "aircraft-chasing".

Done

21) Section 2.1.4, line 155 -- replace "data which were obtained during" with "the measurement data from the" and replace "flights reaching" with "flights that reached".

Done

22) Section 2.1.5, line 159 -- insert "middle and" before "lower troposphere", since Jungfraujoch is (in my opinion) sampling above the lower troposphere. Also change "we used two data sets" to "we also analyse two additional aerosol composition measurement datasets" and replace "low" with "lower".

Done

23) Section 2.1.5, line 160 -- replace "during NETCARE" with "during the NETCARE field campaign".

Done

24) Section 2.1.5, line 164 -- Improve the start of this sentence, changing "During the..." instead to "The other lower altitude dataset is from the mountain-top Jungfraujoch site during the...".

Changed to: "The other lower altitude dataset is from the mountain-top Jungfraujoch site (3600 m a.s.l.) where a single particle mass spectrometer was operated during the INUIT-JFJ (Ice Nucleation Research Unit Jungfraujoch) campaign in January and February 2017."

25) Section 2.2.1, line 175 -- Replace "has been described" with "is described".

Done

26) Section 2.2.1, line 180 -- Replace "Having passed the aerodynamic lens" with "Having passed through an aerodynamic lens", insert "then" before "accelerated" and change "the vacuum chamber" to "a vacuum chamber".

Done

27) Section 2.2.1, line 186 -- Suggest to delete "to the ALABAMA".

Done

28) Section 2.2.1, line 187 -- Suggest to insert "to this paper" after "supplement".

Done

29) Section 2.2.1, line 188 -- Suggest to replace "we include here a subset of" with "we analyse only the measurements from the"

Done

30) Section 2.2.1, line 189 -- Insert "(i.e. where .....)" after "reached the stratosphere" to clarify the criterion that was used for this.

This was done by inspecting the temperature profile of each flight. Only those three flights where a temperature minimum indicated that the tropopause was crossed were selected. We added ", as was inferred from the temperature profiles."

31) Section 2.2.2, line 200 -- replace "briefly reviewed" with "also described".

Done

32) Section 2.2.2, line 200 -- the acronym "CPI" should be spelt out here as "constant pressure inlet" since it is its first use. Note that cloud particle imager also has the same three-letter-acronym.

CPI is first spelled out in Section 2.2.1. We know that the cloud particle imager uses the same acronym, but the constant pressure inlet is published in the meantime by Molleker et al. (2020) in Atmospheric Measurement Techniques (we updated the reference).

33) Section 2.2.2, line 209 -- move "during the StratoClim measurements" to the end of the sentence, as this is more of a clarifying term, i.e. make the sentence instead say "... on particles was about 40\% at diameters around 500nm during the StratoClim measurements".

We moved "during the StratoClim measurements" to the end of the sentence, i.e., after "… and below 5 % above 2000 nm".

34) Caption to Table 1 (line 216) -- change "Overview on the UTLS data sets" to
"Overview of the 5 different aerosol composition measurement datasets".

Changed to "Overview of the 5 different aerosol composition measurement datasets from the UTLS used in this study", because we want to note that these are only the UTLS datasets, not the lower altitude datasets.

35) Table 1 -- Given the issue with these measurements all having a lower frequency (higher wavelength) laser, add a row giving the wavelength used here. Even though these (I think) are all the same at 266nm, it's important to state these here so the reader can easily scan that Table to find that information.

Done

36) Section 2.2.3, line 221 -- insert "5 UTLS" before "campaigns were analyzed".
Also, since this is a European journal, please change all instances of "analyzed" instead to "analysed".

Changed as suggested

38) Section 2.2.3, line 233-234 -- It is really great that the analysis has done this analysis to understand the variations with these different metrics, and the rationale for doing so should be stated. So please change the start of this sentence from "Histograms of..." to "To enable to understand the different origin of the meteoric-signature particles, meteorological re-analysis data was combined with the measurements, with histograms of...", deleting "were" before "calculated" and replace "cluster as function of" with "cluster, as a function of".

Changed as suggested

39) Section 2.2.4, line 238 -- Suggest to improve the start of this 1st para of this section, replacing "The relation of " with "The steep vertical gradients in", and add "across the tropopause, means that correlating with measurements or re-analysis of these species" before "can be used". Then also replacing
"potential tropospheric influence" with "previous tropospheric influence".

Changed to "The steep vertical gradients in water vapor (H2O) and ozone (O3) across the tropopause means that correlations of measurements or re-analysis of these species can be used to investigate the previous tropospheric influence of a stratospheric air mass."

40) Section 2.2.4, lines 239-240 -- the use of the word "tracer" is potentially confusing (e.g. modellers use the word tracer as abbreviation for "trace species"). I suggest with the re-wording in point 39), this sentence can actually be deleted.

The sentence was deleted

41) Section 2.2.4, line 241 -- replace "These measurements are briefly..." with "These additional measurement datasets are briefly..."

Done

42) Section 2.2.4, lines 245, 246 and 247 -- hyphenate these 3 instances of "forward facing" instead to "forward-facing".

Done

43) Section 2.2.4, line 252 -- insert ", whose detection method is" before "based on"

Done

44) Section 2.2.4, line 253 -- replace "of SHARC" with "of the SHARC hygrometer".

Done

45) Section 2.2.4, line 254 -- replace "Monitor" with "monitor"

Done

46) Section 2.2.4, line 256 -- replace "whatever" with "whichever".

Done

47) Section 2.2.4, line 256 -- replace "with an" with "which has an".

Done, assuming that you meant line 259

48) Section 2.2.4, line 265 -- delete "range up to the extreme conditions" and change "at a height of 20km" to "up to a height of ~20km".

Done

49) Section 2.3, line 272 -- insert "for stratifying the data (e.g. the histograms in section 2.2.3)" before "were derived" and replace "using" with "from the".

Done

50) Section 2.3, line 275 -- replace "first lapse rate tropopause" with "lowest altitude negative lapse rate" or some other more precise term.

Done

51) Section 3.1 -- line 279 -- Suggest to replace "Distinct particle type" with "Meteoric-signature particle type" to be more scientifically descriptive.

At this point in our argumentation, we did not draw the conclusion yet that these particles are of meteoric origin. This conclusion is drawn later in section 3.3. Thus we would prefer to leave the section heading as it is.

52) Figure 2 -- as per the main issue I am asking the authors to reply to, there is a question as to the composition of the particles whose spectra do not show any Fe and Mg peaks. The article needs to show the equivalent mean spectra for the non-meteoric-signature particles (ideally separated also into those with carbonaceous and those without carbonaceous, as in Murphy et al., 2007). This should be shown either in additional panels of this Figure 2 or as an additional Figure in the Supplementary Material.

We added to Figure 2 the mean spectra of all stratospheric particles without the meteoric signature. A further separation into different particles types (such as with and without carbonaceous signature) will be subject of upcoming publications that focus on the composition of UTLS aerosol under the influence of Asian and African monsoon.

53) Section 3.1 -- line 291 -- Replace "Further cations include" with "Additional minor cation peaks include"

Done

54) Section 3.1 -- line 292 -- suggest to replace "minor signals" with "trace signals"

Done

55) Section 3.1, lines 294-295 -- insert "the" before "two aircraft missions", insert "with the ERICA" after "missions" and delete "namely".

During CAFE-Africa we used the ALABAMA. Thus, we changed the sentence to "…from the aircraft mission StratoClim 2017 with the ERICA and from the aircraft mission CAFE-Africa 2018 with the ALABAMA".

56) Section 3.1, line 295 -- the word "spectra" is plural but here the term is referring to the mean of the spectra, which is singular, so the word "spectrum" should be used instead of "spectra" in this instance. Also delete "obtained" and insert "the" before "18668 measurements".

Done

57) Section 3.1, line 296 -- replace "during the StratoClim campaign" with simply "during StratoClim", and since the word "spectrum" is used, then the word "look" should be replaced with "looks". The word "compared"

can also be deleted on this line and "the mean mass spectra obtained" replaced with the word "that", also inserting "the" before "3310" and replacing "made during the CAFE-Africa 2018 campaign" with simply "during CAFE-Africa 2018". Those changes make the text much easier to read.

Done. As we also changed Fig. 2 to show only stratospheric particles, the numbers have slightly changed. The sentence now reads: "The mean mass spectrum from the 18421 measurements made during StratoClim 2017 looks remarkably similar to that from the 2882 measurements made during CAFE-Africa 2018."

58) Section 3.1, line 302 -- use the abbreviations Fe and Mg for iron and magnesium on this line and replace "binned" with the more scientific term "stratified".

Done

59) Section 3.1, lines 303-304 -- this sentence beginning "For each bin" can be deleted -- the information there is obvious and just makes this paragraph more difficult to read.

Done

60) In addition to deleting that sentence on lines 303-304, the text after that can be tacked onto the end of the first sentence in that paragraph as ", with bin sizes of ...."

Done

61) Section 3.1, lines 307-309 -- this sentence beginning "It has to be emphasized" can (in my opinion) be deleted -- that is obvious, and the text already gives the total number of particles in the previous sentence, so the reader will have those numbers in their mind already. I think it makes this sentence much easier to read if you simply delete this sentence (the reader will understand that to be the case already).

This sentence is needed to introduce the following statements on the problem with the detection of the pure sulfuric acid particles. Thus, we prefer to keep it.

62) Section 3.1, lines 311-312 -- again, use Fe and Mg abbreviations here rather than the words iron and magnesium. But more importantly this sentence needs to be much clearer how much of an effect this value is. Since Murphy et al. (2007) PALMS measurements, which have the lower wavelength (higher frequency, i.e. higher power) laser, and therefore do sample the pure sulfuric particles, show that these pure sulphuric particles represent only about 10% of the particles. So I think you can say here that the under-sampling of the pure sulphuric particles will not have a significant effect on the fractions given -- and that the reader should be confident in these numbers.

We changed to: "Thus, the fraction of the Fe and Mg particle type given here represents an upper limit and may be overestimated by about 10 – 30 %, because pure sulfuric acid particles are not taken into account. This is discussed in more detail in Section 3.5."

63) Section 3.1, line 324 -- replace "these particles" with "the meteoric-signature particles".

As stated before, the conclusion "meteoric-signature" has not been drawn yet, so we replace by "the Fe and Mg containing particles"

64) Section 3.1, line 330 -- you've written "tropopause" but you mean "troposphere" here -- please correct that. Also insert "often" before "defined via the..." and suggest to add "(known as the thermal tropopause or cold-point tropopause)" after "lapse rate" and make that be the end of that sentence. Then have that start the next sentence ". The potential vorticity" instead of continuing as , but potential vorticity..."

Changed as suggested

65) Section 3.1, line 331 -- I'd suggest "better indicator" rather than "good indicator" -- I think the dynamical tropopause would be the preferred metric if both were available. And please also put the words "dynamical tropopause" in inverted-commas in the manuscript, also changing the preceding words from "indicator for the" instead to ", representing a " so that the sentence is introducing this term.

We hope that this is what the reviewer means:
"The potential vorticity has been found to be a better indicator, representing a "dynamical tropopause" in the extratropics"

66) Section 3.1, lines 337-338 -- reword "during StratoClim 2017 which took place over the AMA" instead to "during the StratoClim 2017 flights sampling above the AMA".

Done

67) Section 3.2, line 351 -- replace "inserted" with "added to the Figure".

Done

68) Section 3.2, lines 352-353 -- please state what time-interval for the individual measurements (across which this median and quartiles are taken).

It is not clear to us what is meant by this comment. The thermal tropopause values were taken from the ECMWF-interpolations along each flight path. Thus, time and lat-lon coordinates correspond to the flights. Otherwise, with respect to the in-situ measurements: All data were taken in their original time resolution (typically 1 second, sometimes 5 seconds) and the median and quartiles were calculated from all values in a certain latitude and temperature bin.

69) Section 3.2, line 353 -- insert "range for the dynamical tropopause is shown, from" before "a 2 PVU and a 5 PVU surface" -- deleting the "a" and replaceing "and" with "to" -- i.e. changing that to be "range for the dynamical tropopause from 2 PVU to 5 PVU".

Changed as suggested

70) Section 3.2, Figure 4 -- in the legends delete the text "with quartiles" -- that can go in the caption to the Figure. Having it in the legend obscures some of the yellow parts of the data, and it would be better then to have the smaller box

and seeing more of the data.

Changed as suggested

71) Section 3.5, line 482 -- Insert the word "accurately" after estimate, and replace "an absolute" with "the absolute".

Done

72) Section 3.5, lines 486-487 -- as per my major comments at the start of this review, this sentence needs to be changed -- it's not appropriate to write "if pure sulfuric acid aerosol particles existed in the air". It's clear from the PALMS measurements that only about 10-20 \% (at most 40%) of particles in the campaigns analysed in Murphy et al. (2007) were of this pure sulphuric particle nature. And you should add a sentence here stating these percentages so that the reader can know that at least two-thirds of the particles (probably more) are being sampled by the 266nm laser used by the ALABAMA and ERICA instruments. That way the reader can know that it is only a relatively small-to-moderate fraction of the particles that might be being missed in these measurements.

We have changed the beginning of Section 3.5 as already detailed above.

73) Section 3.5, line 490 -- insert "mid and" before "lower".

Done

74) Section 3.6, line 590 -- give the range of percentage occurrence that you mean by "was very low". At Jungfraujoch this is 0.2 to 1\%, whereas in the Canadian Arctic the value is much lower. Better to give the corresponding values here.

We added "(0.0025 – 1%)" after "very low"

75) Section 4, lines 621-622 -- Suggest to replace "From previous" with "Consistent with " and insert "aerosol composition measurement" after "previous stratospheric" and replace "it was concluded" with "it is concluded".

Done

76) Section 4, line 626 -- with this being the Discussion and conclusions section, better here not to use the MSP acronym, instead give the words, replacing "MSP particles" with "meteoric smoke particles".

Done

77) Section 4, line 637 -- replace "so this altitude" with "with this altitude", then also replacing "refers to" instead to "referring only to".

Done

78) Supplementary Material -- Introduction, 2nd line (1st sentence) Insert "shown in the main article, to enable its" before "interpretation", and insert "to be scrutinised transparently" after "interpretation.

Done

79) Supplementary Material -- Introduction, 2nd line (2nd sentence) Replace "It includes the clustering parameters...." with "Firstly, the clustering methodology is explained in more detail, with the clustering parameters...", also replacing "evaluation and the uncertainty" with "evaluation, and an associated uncertainty", replacing "estimation" with "estimated".

Done

80) Supplementary Material -- Introduction, 3rd line (3rd sentence) The text "Individual clusters of particles are displayed (S2)" needs to be changed because the Figures S1 to S5 show mean spectra not individual spectra. Also, the vertical profiles of the meteoric fraction are also shown in those Figures. So, replace that text instead with "Secondly (S2), the mean mass spectra and vertical profile of the meteoric-particle abundance fractions for each of the 5 UTLS campaigns are shown in Figures S1 to S5."

Done

81) Supplementary Material -- Introduction, 4th line (4th sentence) Insert "each of" before "the individual" and replace "mission" with "missions", adding "(Figure S6) after "in S3".

Done

82) Supplementary Material -- Introduction, 4th line (5th sentence) Insert "(Figure S7)" before "shows the O3-H2O"...

Done

83) Supplementary Material -- Introduction, 10th line (penultimate sentence in this section) Replace "present in" with "presented in".

Done

84) Supplementary Material -- Introduction, 11th line (final sentence in this section) Replace "SectionS10" with "Section S10", delete "the" after "explains", insert "changing" before "the threshold" and replace "was derived" with "affects the stratospheric proportions presented".

Done

85) Supplementary -- Clustering algorithm, lines 6 & 7 Replace "chose as distance metric" with "used for the distance metric" and replace "spectra): a Pearson..." with "spectra), with a perfect Pearson...", then putting "r=1" in brackets, and changing "means that" to "meaning that".

Changed to: "Linear correlation was used for the distance metric (defining "similarity" of the spectra), with a perfect Pearson correlation (r = 1) meaning that two spectra are identical."

86) Supplementary -- Clustering algorithm, final sentence Replace "stopping" with "convergence".

Done

87) Supplementary -- Variation of clustering parameters, line 6 Replace "particles containing" with "particles identified to contain" and correct "meteorological material" with "meteoric material".

Done

88) Supplementary -- Section S2 -- insert "each of" before "the five".

Done

89) Supplementary -- Section S3 -- insert "each of" before "the five" and replace "mission. All data were merged to" instead to "missions, these data merged to".

Done

90) Supplementary -- Section S8 -- line 8 of the text. Insert "when the refractive index for stratospheric aerosol is used".

Done

91) Supplementary -- Section S8 -- line 9 of the text. Delete "size channel with" and replace "corresponds to" with "increases to", deleting "for stratospheric aerosol particles".

Done

92) Supplementary -- Section S9 -- line 3 of the text. Insert "(NCEP meteorological re-analysis, Saha et al., 2010)" after "0.5 degree data set".

Done

93) Supplementary -- Section S9 -- line 5 of the text. Insert ", with" before "27 trajectories".

Done

94) Supplementary -- Section S9 -- line 6 of the text. Replace "binned in altitude and latitude bins and" with "stratified into altitude and latitude bins and"

Done

95) Supplementary -- Section S10 -- line 1 of the text. Replace "recorded" with "measured".

Done

96) Supplementary -- Section S10 -- line 2 of the text. Replace "We used" with "To test the sensitivity of the calculations, we used"

Done

97) Supplementary -- Section S10 -- line 5 of the text. Replace "as a threshold" with "as the threshold".

Done

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