

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 interpretations 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.

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.

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.

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.

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)?

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.

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.

It sounds like the authors are explaining that only a small proportion of the non-meteoritic-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.

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.

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.

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

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)".

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 Jungfraujoch 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."

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.

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.

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."

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 Jungfrauoch, and smaller abundances (0.0x-0.0y %) from 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 addition 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.

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

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 sentence 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.

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.

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

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.

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.

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.

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.

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

- 17) Introduction, line 116 -- Delete "summer" from the "Tropics/sub-tropics" because this seasonal variation is not relevant here.
- 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.)".
- 19) Section 2.1.1, line 133 -- suggest to insert ", the full dataset from" before "which are included".
- 20) Section 2.1.3, line 147 -- hyphenate "aircraft chasing" to "aircraft-chasing".
- 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".
- 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".
- 23) Section 2.1.5, line 160 -- replace "during NETCARE" with "during the NETCARE field campaign".
- 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...".
- 25) Section 2.2.1, line 175 -- Replace "has been described" with "is described".
- 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".
- 27) Section 2.2.1, line 186 -- Suggest to delete "to the ALABAMA".
- 28) Section 2.2.1, line 187 -- Suggest to insert "to this paper" after "supplement".
- 29) Section 2.2.1, line 188 -- Suggest to replace "we include here a subset of" with "we analyse only the measurements from the"
- 30) Section 2.2.1, line 189 -- Insert "(i.e. where .....)" after "reached the stratosphere" to clarify the criterion that was used for this.
- 31) Section 2.2.2, line 200 -- replace "briefly reviewed" with "also described".
- 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.
- 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".
- 34) Caption to Table 1 (line 216) -- change "Overview on the UTLs data sets" to "Overview of the 5 different aerosol composition measurement 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.
- 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".
- 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".
- 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".
- 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.
- 41) Section 2.2.4, line 241 -- replace "These measurements are briefly..." with "These additional measurement datasets are briefly..."
- 42) Section 2.2.4, lines 245, 246 and 247 -- hyphenate these 3 instances of "forward facing" instead to "forward-facing".
- 43) Section 2.2.4, line 252 -- insert ", whose detection method is" before "based on"
- 44) Section 2.2.4, line 253 -- replace "of SHARC" with "of the SHARC hygrometer".
- 45) Section 2.2.4, line 254 -- replace "Monitor" with "monitor"
- 46) Section 2.2.4, line 256 -- replace "whatever" with "whichever".
- 47) Section 2.2.4, line 256 -- replace "with an" with "which has an".
- 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".
- 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".
- 50) Section 2.3, line 275 -- replace "first lapse rate tropopause" with "lowest altitude negative lapse rate" or some other more precise term.
- 51) Section 3.1 -- line 279 -- Suggest to replace "Distinct particle type" with "Meteoric-signature particle type" to be more scientifically descriptive.
- 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.

- 53) Section 3.1 -- line 291 -- Replace "Further cations include" with "Additional minor cation peaks include"
- 54) Section 3.1 -- line 292 -- suggest to replace "minor signals" with "trace signals"
- 55) Section 3.1, lines 294-295 -- insert "the" before "two aircraft missions", insert "with the ERICA" after "missions" and delete "namely".
- 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".
- 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.
- 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".
- 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.
- 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 ...."
- 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).
- 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.
- 63) Section 3.1, line 324 -- replace "these particles" with "the meteoric-signature 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..."

- 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.
- 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".
- 67) Section 3.2, line 351 -- replace "inserted" with "added to the Figure".
- 68) Section 3.2, lines 352-353 -- please state what time-interval for the individual measurements (across which this median and quartiles are taken).
- 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 replacing "and" with "to" -- i.e. changing that to be "range for the dynamical tropopause from 2 PVU to 5 PVU".
- 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.
- 71) Section 3.5, line 482 -- Insert the word "accurately" after estimate, and replace "an absolute" with "the absolute".
- 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.
- 73) Section 3.5, line 490 -- insert "mid and" before "lower".
- 74) Section 3.6, line 590 -- give the range of percentage occurrence that you mean by "was very low". At Jungfrauoch this is 0.2 to 1\%, whereas in the Canadian Arctic the value is much lower. Better to give the corresponding values here.
- 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".
- 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".
- 77) Section 4, line 637 -- replace "so this altitude" with "with this altitude", then also replacing "refers to" instead to "referring only to".

- 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".
- 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".
- 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."
- 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".
- 82) Supplementary Material -- Introduction, 4th line (5th sentence)  
Insert "(Figure S7)" before "shows the O3-H2O"...
- 83) Supplementary Material -- Introduction, 10th line  
(penultimate sentence in this section) Replace "present in"  
with "presented in".
- 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".
- 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".
- 86) Supplementary -- Clustering algorithm, final sentence  
Replace "stopping" with "convergence".
- 87) Supplementary -- Variation of clustering parameters, line 6  
Replace "particles containing" with "particles identified  
to contain" and correct "meteorological material" with  
"meteoric material".
- 88) Supplementary -- Section S2 -- insert "each of" before  
"the five".
- 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".
- 90) Supplementary -- Section S8 -- line 8 of the text.

Insert "when the refractive index for stratospheric aerosol is used".

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".

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

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

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"

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

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

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

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