**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 Referee #1**

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

Our answers are written in this font style and color.

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

The manuscript submitted presents measurements of meteoric material, identified by its elemental composition, in atmospheric aerosol in the lower stratosphere and in some locations in the troposphere. Observations are presented from a variety of campaigns at a range of latitudes, altitudes and seasons.

Scientific significance:

Measurements of this type in the lower stratosphere are not entirely new, as acknowledged by the authors. However, the statistical analysis made possible by the size of this dataset leads to conclusions regarding trends in the atmospheric abundance of these aerosol which is a new and valuable contribution to the literature. In addition the observation of these particles in the troposphere provides evidence of the occurrence of transport processes which have been previously speculated. I feel that by neglecting aspects of the literature the authors have underestimated the value of their work, and hope to assist in my suggestions below.

Scientific quality:

The scientific approach seems sound and appropriate to the stated aims. In my comments below I suggest several further details which might be usefully discussed.

Presentation quality:

On the whole I find the presentation to be of a standard suitable for publication. I do have some suggestions to improve the readability and effectiveness of the figures. The written English is understandable to me as a native English speaker, though it does use some non-standard (German) sentence structure. I have suggested only typographical language changes.

General comments:

It is my opinion that addressing the following issues will improve the manuscript as presented. I believe these to be minor changes, but acknowledge that some may be more complex than they seem to me. I advise the editor to accept reasonable explanations of why some of my recommendations may not be practical. The most significant change I believe is required is to broaden and better support the scope of the study by including aspects of the literature on meteoric smoke and fragmentation which have been overlooked. This has implications at various points in the manuscript.

Additional literature to discuss:

Bardeen et al. (2008) remains the clearest description of the agglomeration of MSP primary particles in the mesosphere and transport to the stratosphere. This study shows that MSP are formed at a relatively constant rate in the mesosphere, remain too small to sediment and are instead transported into the stratosphere by the downward motion of the polar vortex. This means that it is misleading to state (page 3 line 95) that more MSP are produced from sporadic meteor events than from the constant IDP flux (which dominates the ablated material by mass). In fact both sporadic and constant fluxes feed into the same neutral metal layers which then form MSP. There is therefore a seasonal input of MSP to the polar upper stratosphere, which is then transported to lower latitude.

We agree with the reviewer that our statement on the sporadic events and constant IDP fluxes was incorrect.
We therefore revised this part of the introduction as follows, including also the results by Bardeen et al. (2008) and Brooke et al. (2017). In the first sentence of this part we introduce the term "meteoric material" which encompasses the contribution of IDP, sporadic events, and MSP. Throughout the rest of the manuscript we use only the term "meteoric material".

"The continuous import of submicrometer IDPs, the sporadic events of meteors' disintegration on atmospheric entry, and the meteoric fragments (with radii < 0.5 µm, Brooke et al., 2017) contribute to the atmosphere's load of meteoric material, which becomes incorporated and partially dissolved in acidic aerosols (e.g. of $HNO_3$ and/or $H_2SO_4$ at different dilutions with $H_2O$). Bardeen et al. (2008) investigated ablated meteoric material by means of coupled general circulation model and sectional microphysics model simulations. Due to a mesospheric meridional circulation, as Bardeen et al. (2008) revealed, the re-nucleated meteoric ablation material is transported towards the respective winter pole where it subsides within the polar vortex to stratospheric altitudes. According to the investigations of Dhomse et al. (2013), the nanoparticles released at upper mesospheric altitudes (corresponding to MSP, which are produced by ablation and recombination in the upper atmosphere) reside for about four years in the atmosphere until they are deposited on the surface. The same simulations (Dhomse et al., 2013) predicted the strongest deposition of meteoric ablation material at mid-latitudes with a substantially (~ 15 times) higher efficiency over Greenland than in Antarctica."

Brooke et al. (2017) improved on this work by including interactions of MSP particles with atmospheric sulfate. This study focussed on the difficult task of reproducing measurements of meteoric metals in ice cores, as referenced in the current manuscript.
Brooke et al. (2017) concluded that additional input of meteoric material to the high latitude troposphere was needed, since only a crude treatment of transport in sedimenting large PSC aerosol was able to approach the values measured in the ice cores. The present study, particularly the tropospheric results, represents a valuable data set for future modelling studies to compare to.

The changes in the introduction as given above do also take into account the results by Brooke et al. 2017.

Brooke et al. (2017) also tracked the likely size of MSP agglomerates through the lower atmosphere to surface deposition (figure S5). They showed, in agreement with Bardeen et al. (2008) at higher altitude, that the concentration of MSP particles above 70 nm radius is rather low. This suggests that the particles detected in the present study, with a lower limit of 200 nm diameter, are too large to be MSPs. The size and concentration of fragmented meteor particles is at present unconstrained, however recent publications have suggested that interplanetary dust particles smaller than several hundred nm are rather robust (Mannel et al., 2019), so it is likely that meteoric fragments are large enough to be detected here.

This is a misunderstanding. As we emphasized in our manuscript, the analysed particles consist of meteoric material dissolved in (or possibly coated by) sulfuric acid. All particles that show the meteoric signature (Mg and Fe) show a large sulfuric ($HSO_4^-$) anion signal. However, our method does not allow us to derive the mass fraction of the meteoric material in the particles. Thus, we do not know the original size of the initial MSP that is dissolved in such a $H_2SO_4$ particle of a few hundred nm in diameter.
Therefore, as already mentioned above, we prefer to use the term "meteoric material" for the detected particles by our method.

Dhomse et al. (2013) showed that the residence time of meteoric material transported through the atmosphere as MSP is several years. This is counter to the author's conclusion that "one would therefore expect to find a higher abundance of meteoric particles in the lower stratosphere at high latitudes during late winter and early spring".

But, as you mentioned above, the study by Bardeen et al. (2008) shows "…that MSP are formed at a relatively constant rate in the mesosphere, remain too small to sediment and are instead transported into the stratosphere by the downward motion of the polar vortex".
Thus, we should expect a higher abundance of meteoric material in the outflow of the polar vortex.

We clarified the paragraph in the conclusions section:
"Downward transport of meteoric smoke particles from the mesosphere into the lowermost stratosphere occurs efficiently in the polar vortex (Curtius et al., 2005; Weigel et al., 2014). These papers show that high altitude aircraft measurements demonstrate there is a higher proportion of refractory particles (60-70%) within the wintertime polar vortex, and one would therefore expect to find a higher abundance of meteoric particles in the lower stratosphere at high latitudes during late winter and early spring than at lower extra-tropical latitudes and than in other seasons. This expectation would largely agree with (1) the results by Dhomse et al. (2013), who predicted a more effective (by a factor of ~15) deposition of meteoric ablation material over Greenland than in Antarctica, and (2) the works of Bardeen et al. (2008) and Brooke et al. (2017), according to which the meteoric ablation material most effectively subsides to stratospheric altitudes within the polar winter vortex. This is not confirmed by our observations: Although two mid-latitude campaigns (ML-CIRRUS and ND-MAX/ECLIF-2) were conducted between January and April, we observe the same fraction of meteoric particles at the same ozone levels in the lower stratosphere (Fig. 3) during all campaigns, regardless of latitude and season. "

In addition, the theory that meteoric material leads to nucleation, growth and sedimentation of PSC particles suggests that late winter polar stratosphere may be depleted in meteoric material.

We disagree with this statement. PSCs form at altitudes between around 16 and 24 km, and their sedimentation leads to a re-distribution of, e.g., odd nitrogen to the lower stratosphere (12 – 14 km) where the PSC particles evaporate. For example, Hübler et al. (1990) and Fischer et al. (1997) found elevated $NO_y$ concentrations in the Arctic lower stratosphere at potential temperatures of about 350 K (around 12 km), which is well in the Arctic stratosphere. Therefore, PSC sedimentation would only lead to enhanced downward transport of meteoric material into the lower stratosphere, but not to a removal from the atmosphere by further downward transport into the troposphere.

MSP are likely distributed relatively evenly throughout the stratosphere, with perhaps slightly less presence at lower latitudes (Kremser et al., 2016). However, taking meteoric fragments to have sizes greater than several hundred nm as described above, they would sediment rather rapidly to the lower stratosphere and thus likely also be distributed rather independently of season or latitude. I find the author's conclusion that the meteoric material is evenly distributed to be consistent with current theory of both MSPs and fragments.

As we emphasized above, we can't conclude from our data what the origin of the meteoric material is. We therefore clarified in the conclusions section:

"Our observations of particles with signatures of meteoric material do not clearly indicate the formation history, i.e. whether the material originates from meteoric disintegration by ablation (MSP), fragmentation (MF) or from interplanetary dust particles (IDP), since the meteoric material is at least partially dissolved in sulfuric acid."

We removed the following sentence: "However, the high $H_2SO_4$ content of all detected meteoric particles and the uniform mass spectra suggest that MSPs dissolved in sulfuric acid are the most likely particle source."

The main text of the manuscript currently presents the mass spectra of the detected particles as remarkably reproducible, with the exception that the mass 56 peak is missing in the CAFE-Africa campaign. However looking at the spectra presented in the supplementary material, there is significant variability between clusters identified as meteoric.
The ratio of Mg to Fe, and also the presence or absence of other metals seems rather variable between several meteoric clusters. Specifically: mass 39-41 (39K+, also MgO+ and / or 40Ca+ as assigned by Cziczo et al. (2001)) and mass 27 (Al+). It would be interesting to know if this is an instrumentation issue. Carrillo-Sánchez et al. (2016) discuss the differing elemental composition of sources of interplanetary dust. Variability in the composition of the detected aerosol may also be evidence that the detected particles are variable fragments, rather than MSP, since the latter are agglomerates of many nanoscale particles and should therefore have reproducible composition. Previous works by some of the current authors, using steady state concentration approximations, have produced some of the highest estimates of the meteoric flux to the Earth, on the order of hundreds of tons per day (Weigel et al., 2014;Curtius et al., 2005). This, in comparison to modelling of atmospheric processes comparing the ablated amount of <50 tons per day (Carrillo-Sánchez et al., 2020), suggests that aircraft in the stratosphere are able to observe a portion of the unablated input of meteoric material to the Earth's atmosphere. It would be interesting to know whether the observations presented in this work support this conclusion. If so, then based on this and earlier comments I think the authors should review their conclusion that their detected particles could be either MSP or fragments, or both (P27, line 649). Since fragmentation is at present rather poorly constrained, it is difficult to conclusively say that the particles detected here are fragments, but it also seems unlikely that they are MSPs. If these are fragments then the dataset represents a rare constraint on the flux of this type of meteoric material.

We have had the same idea when analyzing the data and checked whether we could find a dependence of the ion ratios Mg/Fe, Al/Fe, K/Fe, and Na/Fe of latitude, altitude, or potential temperature. However, no significant trend effect was observed. Thus, we conclude that this is an instrumental issue. A random variation in the ion ratios due to the ablation ionization process means that the clustering algorithm will result in a certain number of clusters with different ion ratios, and the number of these resulting clusters depends on the number of prescribed clusters. Summarizing, it is not possible to distinguish MSP and fragments from our method, at least not at our current state of knowledge.

Other comments:

The manuscript presents results using several aircraft and a large number of instruments, measured during a variety of field campaigns. Whilst the terms used are clearly defined, I feel that a reader who was not familiar with these campaigns would benefit from the inclusion of a list of abbreviations.

To our opinion, it is sufficient that all acronyms are spelled out at first use. ACP requires that abbreviations "… need to be defined in the abstract and then again at the first instance in the rest of the text": (https://www.atmospheric-chemistry-and-physics.net/submission.html#manuscriptcomposition). A separate acronym list is not foreseen by ACP.

The manuscript states (p28. Line 665) that "all meteoric particles contained H2SO4, but no other anions like nitrate or organic material." and "This suggests that these particles act similar as pure H2SO4 droplets in the UT with respect to cirrus formation and also in the polar stratosphere with respect to PSC formation." This is unclear. Since nitric acid is only taken up under equilibrium conditions at rather low temperatures in the polar vortex (Clegg et al., 1998), one would not expect to see nitrate signal from these particles with the possible exception of the ND-MAX data, in addition they would likely undergo significant change before the formation of PSC. For upper tropospheric cloud this may be an important observation since concentrated H2SO4 tends to be extremely hygroscopic, meaning that these particles might make extremely effective CCN. On the other hand concentrated H2SO4 is rather viscous, which may limit its ability to take up water (Price et al., 2015). It is unclear to my what the authors mean by this statement, so I suspect it needs additional clarification.

For PCS formation, we are here referring to the works of Tritscher et al. (2019) and James et al. (2018) that were referenced in the introduction. These studies needed to include "foreign nuclei" into their simulations to reproduce PSC observations. Meteoric particles were suggested to be such "foreign nuclei". However, if the particles containing meteoric material "behave" like $H_2SO_4/H_2O$ droplets due to their small mass fraction of meteoric material (20 nm MSP dissolved in 200 nm $H_2SO_4/H_2O$), this information needs to be added to the model.

With respect to cirrus clouds, laboratory measurements (Saunders et al., 2010) showed that refractory particles consisting of $Fe_2O_3$ and MgO nucleated ice under cirrus conditions. However, if particles of meteoric origin are not present as solid particles but as a dilute solution in $H_2SO_4/H_2O$ droplets, the freezing properties will likely change from heterogeneous to homogeneous freezing. However, we agree that dissolved meteoric material might also alter the viscosity of $H_2SO_4$ under low temperature conditions that has been described by Williams and Long (1995).

We therefore changed the statement to:

"Our data further show that all meteoric particles contained $H_2SO_4$, but no other anions like nitrate or organic material. Thus, from our simultaneous cation and anion measurements we can confirm previous assumptions that Mg and Fe are dissolved in $H_2SO_4$ (Murphy et al., 1998; Cziczo et al., 2001; Murphy et al., 2014). This suggests that these particles act similar as pure $H_2SO_4$ droplets in the UT with respect to cirrus formation, but it is conceivable that dissolved

meteoric material alters the viscosity of $H_2SO_4/H_2O$ droplets which was found to increase at low temperatures (Williams and Long, 1995). With respect to PSC formation in the polar stratosphere, the works by James et al. (2018) and Tritscher et al. (2019) showed that "foreign nuclei" are needed to be included in their simulations to reproduce PSC observations. The finding that meteoric material present as dilute solution in $H_2SO_4/H_2O$ droplets needs to be included in future simulations."

**minor and typographical changes:**

Figure 3: Top left panel says m, should say km.

Thanks for pointing out this mistake, it was corrected.

Since the location of the tropopause is later taken to be a set value for each campaign, could this be indicated with a horizontal bar on the relevant panels?

We don't think that this would be helpful. It would mean adding the 380 K line to the StratoClim 2017 plot. temperature graph, adding a shaded area 2-5 PVU for the 4 extratropical campaigns to the PV graph, and then finally a 150 ppb $O_3$ line in the lowest row to all graphs. We also see from Figure 4 that the thermal tropopause varies as a function of latitude and is therefore not a constant value for the individual campaigns.

Page 14 line 330 should read “boundary between troposphere and stratosphere”

Corrected

P16 line 378 whilst “theta-latitude” is a relatively standard term, I find its use here to be somewhat abrupt. This terminology should be standardised throughout the manuscript.

We introduced "theta-latitude space" at the beginning of section 3.2 and use "theta-latitude space" and "theta-equivalent latitude space" throughout the rest of the text.

P21 line 511 “between” should read “above”

Corrected

P22 line 540 & Fig 8. Description of mixing lines is unclear. Perhaps “lines which are not horizontal or vertical” or “data points with intermediate concentrations of both tracers”?

The definition of mixing lines as "lines connecting the respective mixing ratios of the initial unmixed reservoir air parcels" is taken from the referenced Hoor et al. (2002) publication and thus we would prefer to keep it.

P25 Line 585 change to “particles containing”

Corrected

Supplement:

Page S2 first paragraph. I initially understood this to be describing the method for how the cluster was formed, rather than characteristics of a cluster which resulted from the analysis. This would be clearer if relevant sections of main text were referenced, where each characteristic of the cluster are discussed.

The method after which the clusters were selected to contain "meteoric material" are given at the end of section S1:

"Criteria for selecting a certain cluster as "containing meteoric material" were 1) high cation signals of $Fe^+$ and $Mg^+$ (additionally allowing $Na^+$, $K^+$, $Al^+$), 2) anion signal at $HSO_4^-$ or cation signals at $S^+$, $SO_4^+$, $H_3SO_4^+$, 3) vertical profile showing increasing fractional abundance with increasing altitude, potential temperature, or potential vorticity."

We added a sentence explaining this to Section S2.

Page S4 last paragraph, section S10 should say “latter criterion”.

Corrected

Are both panels in Figure S11 on the same horizontal axis?

Yes, the horizontal axis is the same (0.1 - 2 µm), but the vertical axis is different.

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**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
---
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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**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 comment SC1 by J.-P. Renard

Dear Dr. Renard,

Thank you for your comment to our manuscript. Please find the answers to your comments below in blue color
Changes in the revised manuscript are printed in red

This interesting paper presents a nice analysis of the solid material collected in the stratosphere and assumed to originate from meteorites. Nevertheless, I have some troubles with the content of the paper:

1) How can the authors be sure that the analyzed particles have a non-terrestrial origin? Solid particles originated from Earth during various process, or even produced inside the atmosphere, can have the same chemical elements. Some dynamical processes can lift these particles in the stratosphere.

In fact, we discussed this issue in our manuscript in detail (see Sections 3.2 and 3.3):

We have three major arguments:

1) The fraction of the observed Fe- and Mg containing particles increases with altitude (Fig. 3).
2) The composition (cation mass spectra dominated by Mg and Fe, anion mass spectra by sulfate) excludes other (terrestrial or anthropogenic) sources (other sources show different composition, see Section 3.3).
3) Upward transport of tropospheric air masses into the stratosphere occurs mainly in the tropics. However, our data show that in the tropics (here mainly the Asian Monsoon Anticyclone, AMA), the fraction of the Mg- and Fe-containing particle remains low in the lower stratosphere (Fig. 4). See end of Section 3.2: "The observation that the fraction of the iron and magnesium-dominated particle type increases only above the extratropical tropopause layer or mixing layer (Hoor et al., 2002; Hoor et al., 2004; Pan et al., 2004), i.e. 30 K above the tropopause (Fig. 4 b), indicates that the source for this particle type must be above the tropopause, because otherwise, the upwelling air masses in the AMA would contain this particle type also at lower potential temperatures".

2) Confusion is made by the author all along the paper between meteoritic disintegration and interplanetary dust (IDP). Some of the particles of such size (1-300 µm) could be interplanetary dust grains mainly coming from comets, not particles coming from meteorite disintegration. Also, some of these (large) particles can survive the atmospheric entry, as those found in the Antarctica ices. The author must consider the works done by the teams that collect such particles. Also, interplanetary dust and grains coming from meteorites do not have the same composition as cometary grains. The authors must consider the works done on the composition of cometary grains and interplanetary grains, and not only on the composition of meteorites.

We rewrote the introduction with respect to the subject of IPD and meteoric sources, summarize the different possible contributions under the general term "meteoric material" and use this term from there on in the rest of the paper.

We emphasize that from our data we can not distinguish between the different sources of meteoric material, and clarified that again in the conclusions section of the revised version:

"Our observations of particles with signatures of meteoric material do not clearly indicate the formation history, i.e. whether the material originates from meteoric disintegration by ablation (MSP), fragmentation (MF) or from interplanetary dust particles (IDP), since the meteoric material is at least partially dissolved in sulfuric acid."

3) Since the authors have collected a large number of such grains, they can calculate the total concentration and even the total mass-concentration, and they must verify that these values are consistent with the expected flux of solid material (coming from comets and meteorites) that entry the Earth atmosphere. The author must also consider the concentration of interplanetary dust at Earth level.

Unfortunately, the single particle mass spectrometry technique is not able to provide a mass fraction of certain compounds in one single particle. Thus, we do not know how much mass of meteoric material is contained in the detected particles. For example, a particle may have a diameter of 300 nm and is mainly composed of sulfuric acid, but may contain the meteoric material that originates from one meteor smoke particle (MSP) of a few nm in diameter. It may also be that by collision and coagulation more than one MSP ended up in one stratospheric sulfuric acid-dominated particle.
Thus, our method (together with the fact that our data represent only parts of the lower stratosphere) does not allow us to calculate a mass concentration of extraterrestrial material in the atmosphere.

4) We have discussed the problem of the various origins of solid material in the stratosphere, and of the vertical transport of the particles, in the paper now published in “Atmosphere”: J.-B. Renard, G. Berthet, A- C. Levasseur-Regourd, S. Beresnev, A. Miffre, P. Rairoux, D. Vignelles, F. Jégou, Origins and Spatial Distribution of Non-Pure Sulfate Particles (NSPs) in the Stratosphere Detected by the Balloon-Borne Light Optical Aerosols Counter (LOAC), Atmosphere 2020, 11, 1031; doi:10.3390/atmos11101031.
We discuss the IDP and meteoritic material confusion done in many papers; we present a summary of the properties of the IDP and of cometary material. The authors must be advised that we had submitted a few month ago in ACP a previous version of our paper, but it was rejected by an associated editor that is in the same laboratory as one author of this Schneider et al. paper. Obviously, we are sure this is just a coincidence. Nevertheless, we encourage the authors to consider our work and to clarify their analysis considering the various sources that can exist for the material they have identified.

Thank you for pointing us to your recent paper in Atmosphere. We will refer to the results you obtained from stratospheric balloon-borne measurements in the revised version of our manuscript.

One point we would like to mention is that the particles we are describing (the Mg- and Fe-dominated particles we interpret as meteoric material dissolved in sulfuric acid) represent only a subset of the "none-pure sulfate particles" (NSP) that you describe in your publication. In fact, as also reviewer #2 pointed out (and we discussed in section 3.5), our single particle mass spectrometer has a low detection efficiency for pure sulfuric acid. That means, that

almost all particles detected by laser ablation mass spectrometry (using 266 nm ablation laser wavelength) represent NSP. Thus, many of the particles we observed will have terrestrial origin. But, the focus of our present paper is the stratospheric meridional distribution of particles containing meteoric material. Further publications will analyze the nature and sources of the other particles detected during the individual campaigns.

# Aircraft-based observation of meteoric material in lower stratospheric aerosol particles between 15 and 68°N

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**Abstract.** ~~In this paper w~~We ~~analyze~~analyse aerosol particle composition measurements from five research missions ~~conducted~~ between 2014 and 2018 ~~sampling the upper troposphere and lower stratosphere (UTLS),~~ to assess the meridional extent of particles containing meteoric material in the upper troposphere and lower stratosphere (UTLS). Measurements from the Jungfraujoch mountain-top site and a low-altitude aircraft mission ~~Additional data sets from a ground based study and from a low altitude aircraft mission are used to~~show that ~~confirm the existence of~~ meteoric material is also present within middle and lower tropospheric ~~particles~~aerosol, but within only a very small proportion of particles. For both the UTLS campaigns and the lower/mid-troposphere observations, the measurements were conducted with single ~~Single~~ particle laser ablation ~~techniques~~ mass spectrometers with bipolar ion detection which enabled ~~were used~~ to measure the chemical composition of particles in a ~~size~~ diameter range of approximately 150 nm to 3 µm. The five UTLS aircraft missions cover a latitude range from 15 to 68°N, altitudes up to 21 km, and a potential temperature range from 280 to 480 K. In total, 338 363 single particles were ~~analyze~~analysed, of which 147 338 ~~particles~~ were measured in the stratosphere. Of these total particles, 50 688 were characterized by high abundances of magnesium and~~,~~ iron, ~~and rare iron oxide compounds,~~ together with sulfuric ~~acid~~ions, the vast majority ~~. This particle type was found almost exclusively in the stratosphere~~ (48 610 ~~particles~~) in the stratosphere, and ~~is~~ are interpreted as meteoric material immersed or dissolved within ~~stratospheric~~ sulfuric acid ~~particles~~. 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. Below the tropopause, the observed fraction of the meteoric~~is~~ particle type decrease~~d~~ sharply with 0.2 – 1 % abundance at Jungfraujoch, and smaller abundances

(0.025 – 0.05 %). observed during However, small fractional abundances were observed below 3000 m a.s.l. in the the lower altitude Canadian Arctic aircraft measurementsand also at the Jungfraujoch high altitude station (3600 m a.s.l.). The size distribution of the meteoric-sulfuric particles measured in the UTLS campaigns is consistent with earlier aircraft-based mass spectrometric measurements, with only 5-10 % fractions in the smallest particles detected (200 - 300 nm diameter), but with substantial (> 40 %) abundance-fractions for particles from 300 – 350 nm up to 900 nm in diameter, suggesting sedimentation is the primary loss mechanism.Thus, the removal pathway by sedimentation and/or mixing into the troposphere is confirmed. In the tropical lower stratosphere, only a small fraction (< –10 %) of of the analyzeanalysed particles contained meteoric material. In contrast, in the extratropics the observed fraction of meteoric particles reacheds 20-40 % directly above the tropopause. At potential temperature levels of more than 40 K above the thermal tropopause, particles containing meteoric material were observedfound in much higher relative abundances than near the tropopause and, at these altitudes, occurring at similar abundance-fraction across all latitudes and seasons measured. Above 440 440 K, the observed fraction of meteoric particles is ranges betweenabove 60 60 and 80% at latitudes between 20 and 42°N. Meteoric smoke particles This finding suggests that the meteoric material isare transported from the mesosphere into the stratosphere within the winter polar vortex, and ais re efficiently subsequently distributed towards low latitudes by isentropic mixing, typically below above 440 440 K potential temperature. 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 materialThis process can explain that meteoric material is found in particles of the stratospheric aerosol layer at all latitudes.

## 1 Introduction

Aerosol particles in the upper troposphere/lower stratosphere (UTLS) play an important role in the Earth's radiative budget: Firstly, by direct scattering of sunlight back to space, secondly, by influencing homogeneous and heterogeneous cirrus cloud formation in the upper troposphere (UT). The radiative forcing via aerosol-cloud interaction of ice clouds in the atmosphere was estimated in a model study to be around + 0.27 W $m^{-2}$ (Gettelman et al., 2012). Furthermore, especially under volcanically influenced conditions, aerosol particles in the UTLS provide surfaces for heterogeneous chemical reactions to occur, thereby influencing ozone chemistry (Pitari et al., 2002; von Hobe et al., 2011), and influence the stratospheric circulation due to heating of the stratosphere (Robock, 2000; Kremser et al., 2016). Generally, the dominating sources of aerosol particles and their precursor gases are in the troposphere. Primary emitted particles are for example dust, sea spray, black carbon, and biomass burning particles. Secondary aerosol particles are formed from precursor gases such as organic and sulfur-containing compounds. The sources for both primary and secondary particles can be natural or anthropogenic. In contrast, an exclusively natural source of a certain fraction of atmospheric particles is located outside the Earth's atmosphere, causing an ambling but continuous particle import of cosmic origin (Pruppacher and Klett, 1997). The magnitude of cosmic material entering the Earth's atmosphere is currently estimated to be about 43 ± 14 t $d^{-1}$ (tons per day) (Plane, 2012; Carrillo-Sánchez et al., 2016).

Besides oxygen, major elements of meteoric material are iron (Fe), magnesium (Mg), and silicon (Si), which are found with roughly equal proportions in chondritic meteorites; the most abundant minor elements are carbon (C), sulfur (S), aluminum (Al), sodium (Na), calcium (Ca), and nickel (Ni) (Lodders and Fegley Jr., 1998; Hoppe, 2009; Plane et al., 2015). First detection of magnesium emission lines in the night sky spectrum and the conclusion that at least part of atmospheric magnesium is of meteoric origin were reported by Hicks et al. (1972).

About 8 t $d^{-1}$ of the cosmic dust particles (with diameters between ~ 1 µm and ~300 µm) are completely ablated during entry into the Earth's atmosphere (Plane, 2003; Carrillo-Sánchez et al., 2016). Quenching of evaporated compounds is expected to cause their rapid re-nucleation in the mesosphere to form new particles of the size of a few nanometers, which are commonly referred to as meteoric smoke particles (MSPs) (Saunders et al., 2012; Plane et al., 2015; Hervig et al., 2017). Recent remote-sensing and in-situ measurements in the mesosphere indicated that Fe and Mg are the main constituents of MSPs (Hervig et al., 2012; Rapp et al., 2012). MSPs have been identified to act as ice nuclei for noctilucent clouds in the mesopause region (e.g., Alpers et al., 2001; Gumbel and Megner, 2009; Megner and Gumbel, 2009; Rapp et al., 2010) and therefore, it is suggested that MSPs have an impact on polar mesospheric summer echoes (Rapp and Lübken, 2004; Megner et al., 2006). As MSPs are too small to sediment gravitationally, it is widely assumed that MSPs are drained from the mesosphere into the stratosphere most efficiently due to the air mass subsidence within the polar winter vortex, on a timescale of months (Plumb et al., 2002; Curtius et al., 2005; Megner et al., 2008; Plane, 2012; Saunders et al., 2012; Weigel et al., 2014; Plane et al., 2015; Kremser et al., 2016). The aerosol particles in the stratospheric aerosol layer (Junge et al., 1961; Junge and Manson, 1961; Kremser et al., 2016) consist mainly of sulfuric acid-water ($H_2SO_4$-$H_2O$) droplets (Lazrus et al., 1971; Rosen, 1971; Lazrus and Gandrud, 1974, 1977; Sedlacek et al., 1983; Gandrud et al., 1989; Arnold et al., 1998), but a significant fraction of none-pure sulfate particles has been observed by balloon-borne measurements throughout the stratosphere (Renard et al., 2020). The typical size range of the $H_2SO_4$-$H_2O$~~se~~ droplets ranges between 100 and 200 nm (Plane et al., 2015; Kremser et al., 2016). It is thought that MSPs partially dissolve in the droplets (Murphy et al., 1998; Cziczo et al., 2001; Saunders et al., 2012; Murphy et al., 2014), such that a dilute solution of highly soluble ferrous/ferric sulfate and hydrated magnesium sulfate and silicic acid is formed (Saunders et al., 2012). It was further suggested that silicon and aluminum are present as undissolved granular cores within the droplets (Murphy et al., 2014) which would explain the observations of refractory particles in the Arctic lower stratosphere (Curtius et al., 2005; Weigel et al., 2014).

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 ~~As has recently been shown by~~ Subasinghe et al. (2016), about 95 % of cosmic bodies of sizes greater than 1 mm in diameter undergo fragmentation upon entering the Earth's atmosphere, thereby forming unablated meteoric fragments (MFs) of presumably submicron size. If such fragments ~~we~~are formed at sufficient particle concentrations, these particles may sediment directly into the lower stratosphere. It has been suggested that MFs may play a role in polar stratospheric cloud (PSC) formation, thereby influencing polar ozone destruction (Voigt et al., 2005; James et al., 2018). Satellite-based observations of PSCs in the Arctic were reproduced by model simulations using CLaMS (Chemical Lagrangian Model of the Stratosphere), but only if heterogeneous nucleation of NAT (nitric acid trihydrate, Grooß

et al., 2014) and ice particles (Tritscher et al., 2019) on foreign nuclei were included in the model parameterization. A potential source of the foreign nuclei is meteoric dust (James et al., 2018).

Additionally, certain amounts of cosmic particulate material enter the Earth's atmosphere as Interplanetary Dust Particles (IDPs) which, if smaller than 1 µm in diameter, are too small to experience any ablative altering during atmospheric entry. The origin of IDPs is mainly attributed to collisions of asteroids, sublimation of comets and long-decayed cometary trails (Plane, 2003, 2012). In terms of the size-segregated mass influx of cosmic particles (Plane, 2003, 2012), the contribution of submicrometer sized IDPs to the total atmospheric aerosol particle load is estimated to be small with estimates of about 150 t per year.

The continuous import of submicrometer IDPs, the sporadic events of meteors' disintegration on atmospheric entry, and the meteoric fragments (with radii < 0.5 µm, Brooke et al., 2017) contribute to the atmosphere's load of meteoric material, which becomes incorporated and partially dissolved in acidic aerosols (e.g., of $HNO_3$ and/or $H_2SO_4$ at different dilutions with $H_2O$). Bardeen et al. (2008) investigated ablated meteoric material by means of coupled general circulation model and a sectional microphysics model simulations. Due to a mesospheric meridional circulation, as Bardeen et al. (2008) revealed, the re-nucleated meteoric ablation material is transported towards the respective winter pole where it subsides within the polar vortex to stratospheric altitudes. According to the study by Dhomse et al. (2013), the nanoparticles originally released at upper mesospheric altitudes (corresponding to MSPs, which are produced by ablation and recombination in the upper atmosphere) reside for about four years in the atmosphere until they are deposited on the surface. The same simulations (Dhomse et al., 2013) predicted the strongest deposition of meteoric ablation material at mid-latitudes with a substantially (~ 15 times) higher efficiency over Greenland than in Antarctica.~~However, the import of IDP is likely a continuous process compared to sporadic events of meteoric entries that produce by far more MSP per event than a single IDP. Therefore, an ambling and persistent import of cosmic aerosol particles (by number) should be considered in relationship to the infrequent but then excessively effective ablation/fragmentation events releasing large amounts of MSP and MF in the atmosphere.~~

The existence of particles containing meteoric material in the lower stratosphere has been shown by direct in-situ observations. Mossop (1965) reported on insoluble inclusions found in stratospheric particles sampled at 20 km by the U-2 aircraft and suggested a meteoric origin of these particles. Shedlovsky and Paisley (1966) ~~analyze~~analysed particles sampled by the U-2 aircraft and detected sulfur, iron, sodium, copper, and chromium. More recently, ~~Later,~~ aircraft-based in-situ aerosol mass spectrometry in the tropical and mid-latitude lower stratosphere at altitudes up to 19 km showed a significant fraction of particles containing meteoric material and sulfuric acid (Murphy et al., 1998; Cziczo et al., 2001; Froyd et al., 2009; Murphy et al., 2014). Indirect evidence for the existence of meteoric aerosol particle material in the Arctic lower stratosphere up to 20 km altitude was reported by Curtius et al. (2005) and Weigel et al. (2014). They measured non-volatile particles that were thermally stable on exposure to 250°C and had diameters of 10 nm to a few micrometers. The fraction of these non-volatile particles increased with altitude up to 70% at potential temperature levels between 430 and 500 K. Ebert et al. (2016) report on submicrometer particles collected with a cascade impactor in the Arctic stratosphere during the winters 2010 and 2011 that were ~~analyze~~analysed for their chemical composition and morphology. They found Fe-rich particles, Ca-rich particles,

silicates, silicate/carbon mixed particles and mixed metal particles from different sources, such as meteoric material, space debris and to lower extent terrestrial sources.

Here we report findings from aircraft measurements of aerosol particle composition in the lower stratosphere at different altitudes, latitudes and seasons:

- Western Europe, spring (March-April 2014) and summer (August-September 2018),
- Mediterranean, summer (August-September 2016),
- Tropics/subtropics, ~~summer~~ (July-August 2017 and August 2018),
- North America/Northern Atlantic, winter (January-February 2018),

In all data sets we observed a distinct particle composition type in the lower stratosphere that can be interpreted as particles containing meteoric material, dissolved in or coated by sulfuric acid. We discuss mass spectral composition, size distribution, vertical profiles, latitudinal distribution, and cross-tropopause transport of particles containing meteoric material.

## 2 Measurements and Methods

### 2.1 Field measurements

This study analyses ~~includes~~lower stratospheric and upper tropospheric ~~data obtained~~aerosol composition measurements taken during five aircraft-based field~~research~~ campaigns, together with two additional composition measurements ~~data sets~~ from the middle and lower troposphere (altitudes below 3600 m a.s.l). The individual campaigns are described briefly in the following. The flight tracks of all UTLS research flights included here as well as the locations of the low altitude measurements are depicted in Fig. 1. Overview about the five UTLS aircraft campaigns is provided in Table 1.

#### 2.1.1 ML-CIRRUS

The field campaign ML-CIRRUS (Mid-Latitude Cirrus) was conducted in March and April 2014 out of Oberpfaffenhofen, Germany, using the research aircraft HALO (High Altitude and Long Range Research Aircraft). The objective of ML-CIRRUS was to study cirrus clouds by in-situ and remote sensing methods. Including test flights, a total of 16 flights were carried out. Most of the flight time (in total 88 hours) was spent in the upper troposphere and lower stratosphere. Laser ablation aerosol mass spectrometer data were recorded during 15 flights, the full dataset from which are included in this study. A detailed overview on the mission is given by Voigt et al. (2017).

#### 2.1.2 StratoClim

Two aircraft-based research campaigns were conducted within StratoClim (Stratospheric and upper tropospheric processes for better climate predictions) which is a collaborative research project funded by the European Commission. The first StratoClim campaign took place at Kalamata airport, Greece, in August and September 2016. The aim of the mission was to study

atmospheric composition in the Eastern Mediterranean region, including the remote influence of the Asian monsoon anticyclone (AMA) outflow. Three research flights were conducted. The second StratoClim campaign was a dedicated field activity to investigate the impact of the AMA on the UTLS and took place at the Tribhuvan International Airport of Kathmandu, Nepal, in July and August 2017 (e.g., Höpfner et al., 2019). Eight scientific flights were carried out over Nepal, India and Bangladesh. The flight paths spanned latitudes from 21° N to 27° N and longitudes from 79° E to 90° E (see Fig. 1).

### 2.1.3 ND-MAX/ECLIF-2

The ND-MAX/ECLIF-2 (NASA/DLR-Multidisciplinary Airborne eXperiments/Emission and CLimate Impact of alternative Fuel) mission aimed for the characterization of gaseous and particulate aircraft emissions with a dedicated aircraft ~~–~~chasing field experiment over South-West Germany. For this mission, the installation of instrumentation into the NASA DC-8 aircraft took place at Palmdale, CA, USA. Measurements taken during the ferry flights from Palmdale to Germany on January 13, 2018 and back on February 3 and 4, 2018, were used in this study. These flights reached latitudes up to 68°N (see Fig. 1), longitudes as far as 120° W, and penetrated deep into the winter stratosphere at around 11 km altitude.

### 2.1.4 CAFE-Africa

CAFE-Africa (Chemistry of the Atmosphere Field Experiment in Africa) was conducted with HALO in August 2018 out of Sal on the Cape Verde Islands. The main objective was to study the African monsoon outflow in the upper troposphere over the Atlantic Ocean. This study includes only ~~data which were obtained during~~the measurement data from the three research flights that ~~reaching~~ reached the stratosphere, as was inferred from the temperature profiles. These flights took place on August 15, August 24, and September 07, 2018, the latter being the ferry flight back to Germany. The flight tracks of these three flights are included in Fig. 1.

### 2.1.5 Additional low altitude data sets

To investigate the possible occurrence of meteoric particles in the middle and lower troposphere, we also analyse ~~used~~ two additional aerosol composition measurement data-sets from lower altitudes: One data set was obtained during the NETCARE field campaign (Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments (Abbatt et al., 2019)), conducted in the Arctic out of Resolute Bay (Nunavut, Canada) in July 2014. A single particle mass spectrometer was operated on board the Polar 6 aircraft (Alfred Wegener Institut – Helmholtz Zentrum für Polar- und Meeresforschung) and measured at altitudes between 0 and 3 km. Details of the campaign and the mass spectrometer data are given in Köllner et al. (2017). 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 ~~During the~~ INUIT-JFJ (Ice Nucleation Research Unit Jungfraujoch) campaign in January and February 2017. ~~, a single particle mass spectrometer was operated for ground based measurements on the High Alpine Research Station Jungfraujoch (3600 m a.s.l.).~~ The mass spectrometer data are still unpublished, but details on the campaign can be found in Eriksen Hammer et al. (2018) and Gute et al. (2019).

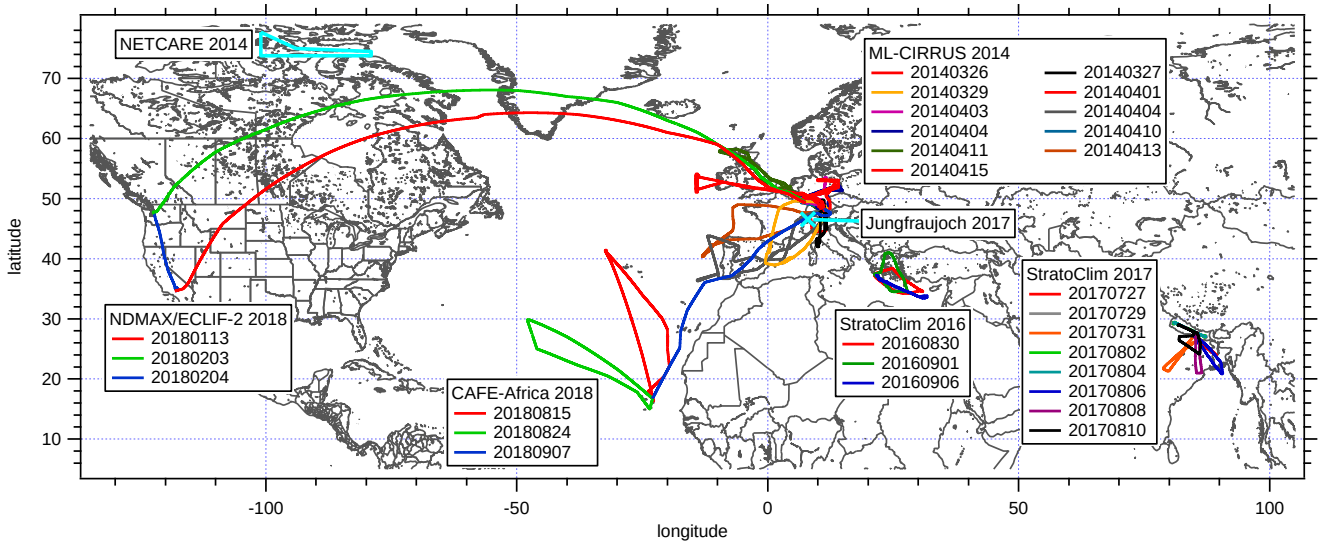



**Figure 1: Map of the flight tracks of all UTLS research flights used in this study. Additionally the locations of the low altitude measurements are indicated: Jungfraujoch (3600 m a.s.l.) and operation range of the NETCARE flights (0 – 3000 m a.s.l.).**

200

### 2.2 Instrumentation and data analysis

#### 2.2.1 Aerosol mass spectrometer operated during ML-CIRRUS and CAFE-Africa

The aircraft-based laser ablation aerosol mass spectrometer (ALABAMA) ~~has been~~is described in detail in Brands et al. (2011), Köllner et al. (2017), and Clemen et al. (2020). Briefly, the ALABAMA is a bipolar ion single particle analysis instrument 205 that samples aerosol particles from ambient air through a constant pressure inlet (CPI, Molleker et al., 2020) and an aerodynamic lens. The sampled particle size range (vacuum aerodynamic diameter $d_{va}$, DeCarlo et al. (2004)) was between about 200 and 1000 nm during ML-CIRRUS and between 200 nm and 3000 nm during CAFE-Africa. Having passed through an ~~the~~ aerodynamic lens, the particles are then accelerated into ~~the~~ a vacuum chamber. The particles are detected by two 405 nm laser diodes and their velocity information is used to determine their vacuum aerodynamic diameter and to trigger a 210 laser shot of the ablation laser (quadrupled Nd:YAG, 266 nm) that hits the particles in the ionization region of the bipolar ion time-of-flight mass spectrometer. During ML-CIRRUS and CAFE-Africa, aerosol particles were sampled through the HALO aerosol submicrometer inlet (HASI, Andreae et al., 2018). The inlet was mounted on the upper side of the fuselage of the aircraft. Inside the aircraft, the sampled aerosol particles were guided through a 2.9 m long stainless steel sampling line with an inner diameter of 5 mm ~~to the ALABAMA~~. The calculated transmission efficiency of this sampling line is shown in the 215 supplement to this paper (Fig. S~~10~~9). During ML-CIRRUS, the ALABAMA was operative during 15 flights and ~~analyze~~analysed more than 24000 ambient aerosol particles (see Table 1). From CAFE-Africa, we analyse only the

measurements from the ~~include here a subset of~~ three flights where HALO reached the stratosphere, as was inferred from the temperature profiles. In these three flights the ALABAMA sampled and ~~analyze~~analysed more than 65000 particles. The higher efficiency and higher upper size cut-off (see above) of the ALABAMA in CAFE-Africa compared to ML-CIRRUS are due to several instrumental improvements such as a new aerodynamic lens system and delayed ion extraction (Clemen et al., 2020). The ALABAMA was also used in the above-mentioned low altitude field campaigns NETCARE and INUIT-JFJ.

### 2.2.2 Aerosol mass spectrometer operated during StratoClim and ND-MAX

The newly developed mass spectrometer ERICA (ERC Instrument for Chemical composition of Aerosols) combines single particle laser ablation and flash vaporization/ionization techniques. It was designed for fully automated operation on the high altitude research aircraft M-55 "Geophysica" during the StratoClim project and was later re-configured to be operated on the NASA DC-8 during the ND-MAX/ECLIF-2 mission. Here we use only data obtained using the laser ablation part of the ERICA (ERICA-LAMS). The basic design is similar to that of the ALABAMA, but since this is a newly developed instrument, it is also described ~~briefly reviewed~~ here. The aerosol particles are sampled via a CPI (Molleker et al., 2020) and an aerodynamic lens designed for PM2.5 (Peck et al., 2016). In the vacuum chamber, the particles are detected by two laser diodes (405 nm) and ablated by a pulsed quadrupled Nd:YAG laser (Quantel Ultra, LUMIBIRD SA, Roubaix, France) emitting at 266 nm. This Nd:YAG laser is operated without a wavelength separator in the laser head and thus emits also a small fraction of the energy in form of the first and second harmonic (1064 and 532 nm). The generated ions are ~~analyze~~analysed in a bipolar ion time-of-flight mass spectrometer (Tofwerk AG, Thun, Switzerland). The size range of the ERICA-LAMS is approximately 100 – 5000 nm ($d_{va}$). Particle size was calibrated using PSL (polystyrene latex) particles with diameters between 80 nm to 5000 nm. The particle detection efficiency at the laser diodes reaches a maximum of about 75% at 400 nm and decreases towards lower and higher diameters. The hit rate, defined as the ratio of recorded particle mass spectra to laser shots on particles was ~~during the StratoClim measurements~~ about 40 % at diameters around 500 nm, between 5 and 10 % below 200 nm, 20 % at 1000 nm, and below 5 % above 2000 nm during the StratoClim measurements. During StratoClim, the ERICA was operated on 11 research flights (three in 2016 and eight in 2017), and the ERICA-LAMS ~~analyze~~analysed about 150 000 single particles (see Table 1). During the three ferry flights conducted in the ND-MAX/ECLIF-2 project that are used here, the ERICA-LAMS recorded more than 98 000 single particle mass spectra.

Table 1. Overview of the five different aerosol composition measurement ~~in the UTLS~~ data-sets from the UTLS used in this study.

| Project | ML-CIRRUS | StratoClim 2016 | StratoClim 2017 | ND-MAX | CAFE-Africa |
|---|---|---|---|---|---|
| Time | Mar – Apr 2014 | Aug – Sep 2016 | Jul – Aug 2017 | Jan - Feb 2018 | Aug – Sep 2018 |
| Measurement region | Western Europe | Eastern Mediterranean | South Asia | U.S. to Europe | Atlantic Ocean |
| Aircraft | HALO (G550) | M-55 Geophysica | M-55 Geophysica | NASA DC-8 | HALO (G550) |
| Instrument | ALABAMA | ERICA | ERICA | ERICA | ALABAMA |
| Ablation laser wavelength | 266 nm | 266 nm | 266 nm | 266 nm | 266 nm |
| No. of flights used in this study | 15 | 3 | 8 | 3 | 3 |
| Altitude range (km) | up to 13.8 km | up to 20.2 km | up to 20.5 km | up to 11 km | up to 14.5 km |
| Theta range (K) | 276 - 387 | 295 - 490 | 310 - 480 | 276 – 340 | 295 - 380 |
| Latitude range (° N) | 36.3 – 57.5 | 33.4 – 41.0 | 20.8 - 29.5 | 34.6 – 68.1 | 15.0 – 48.2 |
| PV range (PVU) | 0 - 10 | 0 – 24 | 0 - 22 | 0 - 8 | 0 - 10 |
| Total number of single particle mass spectra | 24833 | 11709 | 138119 | 98598 | 65104 |
| In stratosphere (PV > 4 PVU) | 6509 | 5092 | 51599 | 73367 | 10771 |
| Number of mass spectra dominated by magnesium and iron | 3140 | 2412 | 18688 | 23138 | 3310 |
| In stratosphere (PV > 4 PVU) | 2986 | 2271 | 18421 | 22050 | 2882 |

### 2.2.3 Single particle mass spectrometer data analysis

The aircraft data sets from all five UTLS campaigns were analysed using a consistent procedure to ensure comparability of the results. First, all data measured during one campaign were merged into one data set per campaign. This resulted in data sets containing individual spectra information of 11 709 particles (StratoClim 2016) to up to 138 119 particles (StratoClim 2017) as given in Table 1. These data sets were clustered separately using a fuzzy c-means algorithm (for a general description see Bezdek et al. (1984) and Hinz et al. (1999); for an ALABAMA-specific description see Roth et al. (2016)), with a pre-selected number of 20 clusters per campaign. Only cations were considered for the clustering algorithm for two reasons: First, during ML-CIRRUS many anion mass spectra were too noisy. Second, the particle type of interest was found to be mainly characterized by the cation mass spectrum, containing magnesium and iron, as explained in the next section. Further clustering details are given in the supplement (Section S1 and Table S1). For quality assurance and uncertainty estimation, the clustering was repeated using different starting conditions and also different algorithms. The results showed only small deviations in the type of clusters and in the numbers of mass spectra attributed to the clusters (supplement, Section S1 and Table S2). Mean mass spectra (anions and cations) were calculated for each cluster and were used for the interpretation of the particle type associated with this cluster. To enable to understand the different origin of the meteoric-signature particles, meteorological re-analysis data was combined with the measurements, with histogramsHistograms of relative particle abundance were calculated for each cluster, as a function of altitude, potential temperature ($\Theta$), and potential vorticity (PV).

The data sets from low altitudes (NETCARE, Jungfraujoch) were treated differently: Here we searched specifically for mass spectra using selected marker ions that were found in the high altitude data. This is explained later in detail (Section 3.7).

### 2.2.4 Auxiliary data

The steep vertical gradients inThe relation of water vapor ($H_2O$) and ozone ($O_3$) across the tropopause means that correlations of measurements or re-analysis of these species can be used to investigate the previouspotential tropospheric influence of a stratospheric air mass. To study the transport of the particles across the tropopause, we use $O_3$ as stratospheric tracer and $H_2O$ as a tropospheric tracer. We use independent particle number concentration and particle size measurements to convert the mass spectrometer data to number concentrations. These additional measurement datasets are briefly explained here.

Water vapour was measured during ML-CIRRUS and StratoClim by the airborne Fast In-situ Stratospheric Hygrometer (FISH). This instrument uses Lyman-alpha photofragment fluorescence and is described in detail by Zöger et al. (1999). The detection limit is reported to be below 0.4 ppmv, the uncertainty was determined to be about 8 – 30% for low $H_2O$ mixing ratios (1 – 4 ppmv) and 6 – 8% between 4 and 1000 ppmv (Meyer et al., 2015). During ML-CIRRUS, FISH sampled the air through a forward--facing inlet mounted on the upper fuselage of the HALO aircraft, whereas during StratoClim, the forward-facing FISH inlet was mounted on the side of the fuselage of the Geophysica aircraft (Afchine et al., 2018). The forward--facing inlet also samples cloud droplets and ice crystals which evaporate in the inlet, such that the FISH measurements refer

to total water. We therefore restricted the data set to non-cloud conditions, by removing the data points where the $H_2O$ saturation ratio was greater than 0.8. During ND-MAX/ECLIF-2, water vapor was measured using the Diode Laser Hygrometer (DLH) of NASA/LaRC (Diskin et al., 2002), which has an uncertainty of 5%. During CAFE-Africa, water vapour was measured by SHARC (Sophisticated Hygrometer for Atmospheric ResearCh), whose detection method is based on direct absorption measurement by a tunable diode laser (TDL) system. The uncertainty of the SHARC hygrometer is 5% or ± 1 ppmv. Ozone was measured during ML-CIRRUS and CAFE-Africa by the Fast Airborne Ozone m~~M~~onitor (FAIRO), whereas during StratoClim, $O_3$ was measured by the Fast Ozone ~~Analyze~~Analyser (FOZAN-II). FAIRO combines a dry chemiluminescence detector (CI-D) with a 2-channel UV photometer. The total uncertainty is 1.5% at 8 Hz or 1.5 ppb, whichever is higher, see Zahn et al. (2012). FOZAN-II is likewise based on a CI-D and has a uncertainty of less than 10%, see Yushkov et al. (1999) and Ulanovsky et al. (2001). During ND-MAX, $O_3$ was measured by the UV photometric Ozone ~~analyze~~analyser TE49 (Thermo Scientific) which has~~ith~~ an uncertainty of 5%.

Aerosol particle size distributions were measured during both HALO missions using an optical particle spectrometer of type Grimm 1.129 "Sky-OPC" which was installed next to the ALABAMA. The Sky-OPC measured the total particle number concentration and size distribution for particles larger than 250 nm (manufacturer calibration) in diameter with a reproducibility of 3%, given by the manufacturer. During the StratoClim campaigns, we used a modified Ultrahigh Sensitive Aerosol Spectrometer (UHSAS-A), with a particle diameter range from 65 nm to 1000 nm. The modifications allowed for an airborne application ~~range up to the extreme conditions~~ in the stratosphere up to~~at~~ a height of 20 km. The measurement uncertainties of the UHSAS were determined to 10% (Mahnke, 2018).

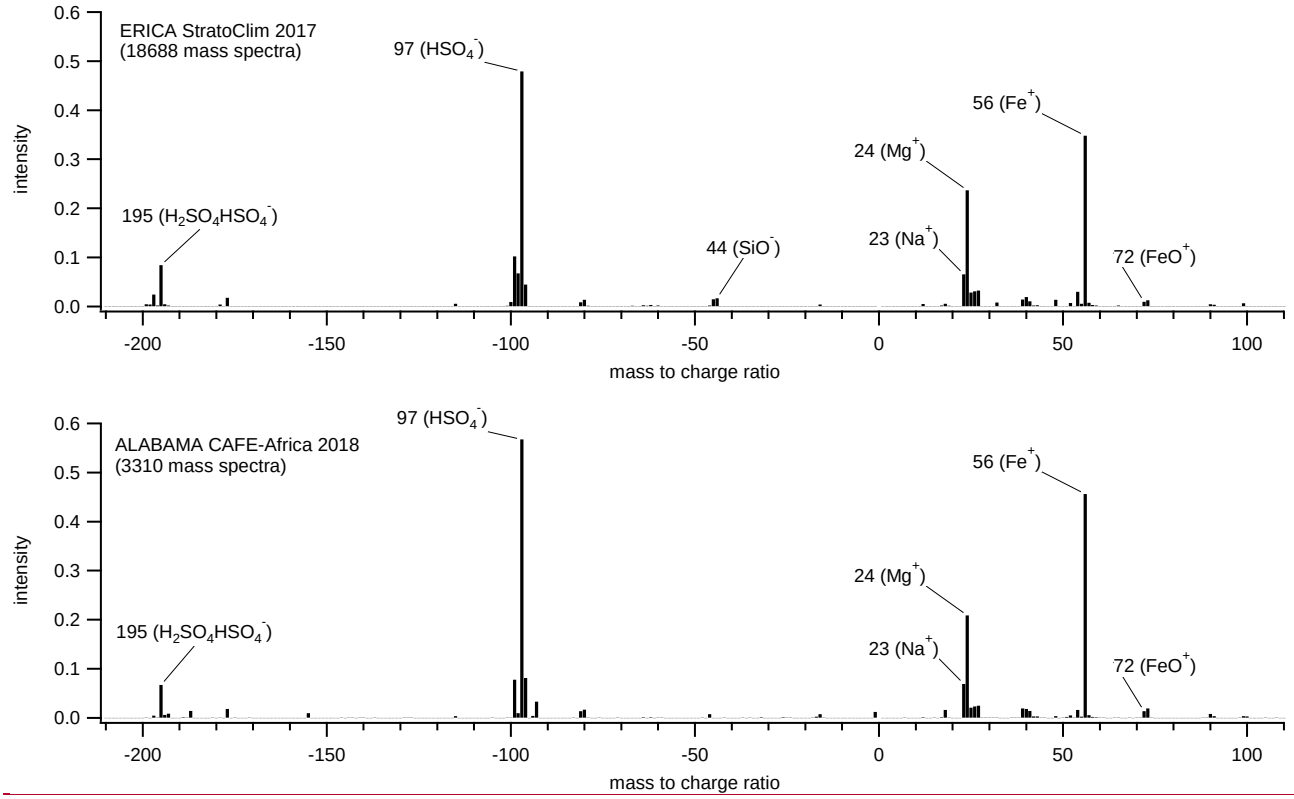
Basic meteorological parameters such as pressure, temperature, as well as aircraft position and altitude were obtained during ML-CIRRUS and CAFE-Africa from the Basic HALO Measurement and Sensor System (BAHAMAS), during StratoClim from the Unit for Connection with the Scientific Equipment (UCSE), and during ND-MAX/ECLIF-2 from the NASA DC-8 facility instrumentation.

### 2.3 Meteorological reanalysis

Meteorological parameters for stratifying the data (e.g., the histograms in section 2.2.3) were derived ~~using~~ from the ERA-Interim reanalysis (Dee et al., 2011) from the European Centre of Medium Range Weather forecast (ECMWF). For meridional characterization we use equivalent latitude (Lary et al., 1995) from ERA-Interim. For vertical coordinate, we use potential vorticity from ERA-Interim and potential temperature derived from observed pressure and temperature data. The location of the thermal tropopause (lowest altitude negative lapse rate~~first lapse rate tropopause~~) in potential temperature coordinates was taken from ERA-Interim.

## 3 Results

### 3.1 Distinct particle type containing magnesium and iron ions


ERICA StratoClim 2017
(18688 mass spectra)
97 (HSO4-)
56 (Fe+)
24 (Mg+)
195 (H2SO4HSO4-)
44 (SiO-)
23 (Na+)
72 (FeO+)
intensity
mass to charge ratio
ALABAMA CAFE-Africa 2018
(3310 mass spectra)
97 (HSO4-)
56 (Fe+)
24 (Mg+)
195 (H2SO4HSO4-)
23 (Na+)
72 (FeO+)
intensity
mass to charge ratio

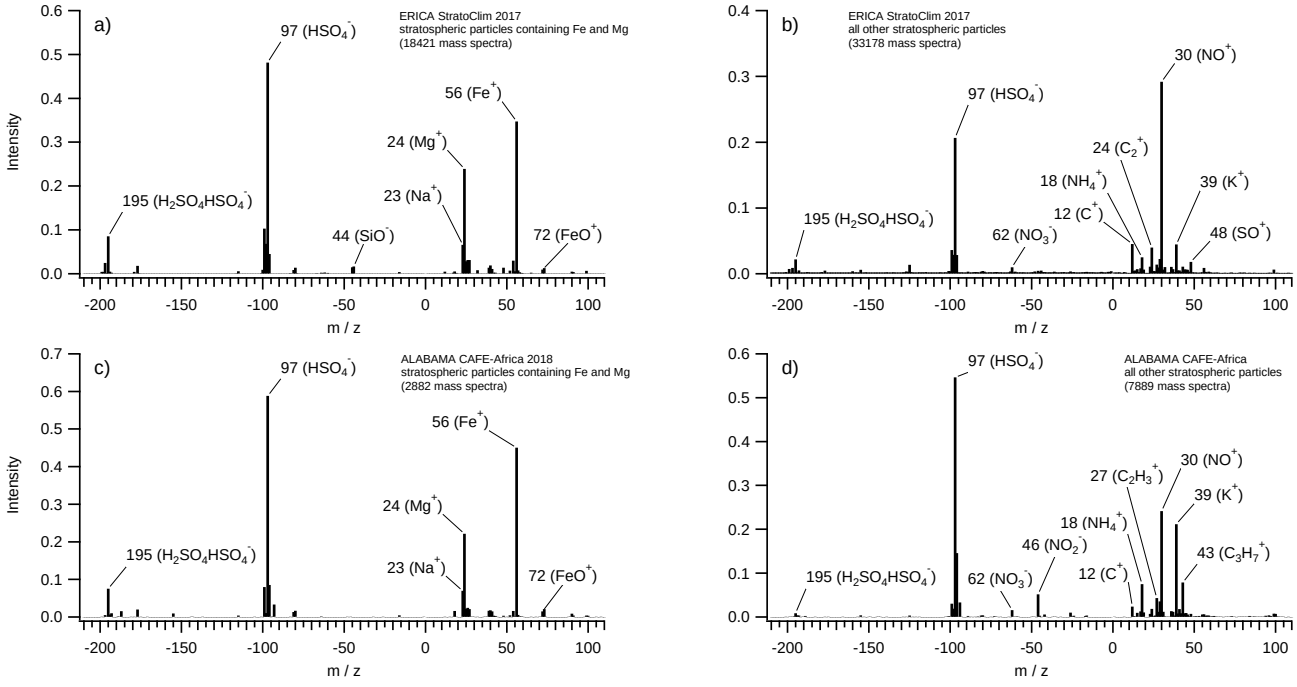

a)
ERICA StratoClim 2017
stratospheric particles containing Fe and Mg
(18421 mass spectra)
97 (HSO4-)
56 (Fe+)
24 (Mg+)
195 (H2SO4HSO4-)
23 (Na+)
44 (SiO-)
72 (FeO+)
Intensity
m / z
b)
ERICA StratoClim 2017
all other stratospheric particles
(33178 mass spectra)
30 (NO+)
97 (HSO4-)
24 (C2+)
18 (NH4+)
39 (K+)
12 (C+)
195 (H2SO4HSO4-)
62 (NO3-)
48 (SO+)
m / z
c)
ALABAMA CAFE-Africa 2018
stratospheric particles containing Fe and Mg
(2882 mass spectra)
97 (HSO4-)
56 (Fe+)
24 (Mg+)
195 (H2SO4HSO4-)
23 (Na+)
72 (FeO+)
Intensity
m / z
d)
ALABAMA CAFE-Africa
all other stratospheric particles
(7889 mass spectra)
97 (HSO4-)
30 (NO+)
27 (C2H3+)
39 (K+)
18 (NH4+)
46 (NO2-)
43 (C3H7+)
195 (H2SO4HSO4-)
62 (NO3-)
12 (C+)
m / z

**Figure 2: Mean mass spectra of stratospheric particles (PV > 4 PVU). Panels a), b): StratoClim 2017 (ERICA); Panels c), d): CAFE-Africa 2018 (ALABAMA). Panels a) and c) show the averaged mass spectra ~~clusters containing~~of particles of which the positive mass spectra are dominated by iron ($Fe^+$, $FeO^+$) and magnesium ($Mg^+$). ~~The upper panel shows the average over 18688 mass spectra recorded by the ERICA during StratoClim 2017, the lower panel shows an average over 3310 ALABAMA mass spectra recorded during CAFE-Africa 2018.~~ The two anion as well as the two cation mass spectra of the Mg and Fe-dominated particles correlate between the instruments with $r^2$ = 0.97~~. The only difference is the detection of $SiO^-$ (m/z 44) by the ERICA~~. Panels b) and d) show the averaged mass spectrum of all other stratospheric particles.**

In all five upper tropospheric and lower stratospheric aircraft data sets, the clustering algorithm yielded a type of mass spectra with a mean cation mass spectrum characterized by high abundance of magnesium ($Mg^+$, m/z 24 for the major isotope, m/z 25 and 26 for the minor isotopes) and iron ($Fe^+$, m/z 56 for the major isotope, m/z 54 for the most abundant minor isotope). Also oxides of Fe ($FeO^+$, m/z 72; $FeOH^+$, m/z 73) were clearly detected. Additional minor cation peaks ~~Further cations~~ include sodium ($Na^+$, m/z 23), aluminum ($Al^+$, m/z 27), as well as ~~minor~~ trace signals of potassium ($K^+$, m/z 39 and 41) and calcium ($Ca^+$, m/z 40). The mean anion mass spectrum contains almost exclusively sulfuric acid ions, as $HSO_4^-$ (m/z 97) and $H_2SO_4HSO_4^-$ (m/z 195). Figure 2 shows the averaged bipolar ion ~~spectra~~ spectra of stratospheric particles (PV > 4 PVU) ~~this particle type~~ from the ~~two~~ aircraft mission~~s, namely~~ StratoClim 2017 with the ERICA and from the aircraft mission CAFE-Africa 2018 with the ALABAMA (panels a) and c)). The mean mass ~~spectra~~ spectrum of the Fe and Mg particle type ~~obtained~~ from the 18421~~668~~ measurements made during ~~the~~ StratoClim 2017 ~~2017 campaign~~ look**s** remarkably similar ~~compared~~ to ~~the mean mass spectra obtained~~that from the 2882~~3310~~ measurements made during ~~the~~ CAFE-Africa 2018 ~~campaign~~. A linear correlation between the mean mass spectra of the Fe and Mg particle type measured during CAFE-Africa and StratoClim yielded an $r^2$ of 0.97 for both the anions and the cations. The only difference is the detection of $SiO^-$ (m/z 44) by ERICA during StratoClim 2017 that was missing from the CAFE-Africa observations. This might be due to the additional emission of 1064 and 532 nm light of the ERICA laser in contrast to the ALABAMA laser, such that the ionization probability of Si-containing compounds is higher in the ERICA than in the ALABAMA. Additionally, panels b) and d) of Figure 2 show the averaged mass spectrum of all other stratospheric particles measured during the two campaigns. These spectra contain anion and cation signals indicating nitrate ($NO_3^-$, $NO_2^-$, $NO^+$) and several ion signals indicating carbonaceous material ($C^+$, $C_2^+$, $C_2H_3^+$, $C_3H_7^+$). The observation that carbonaceous material is found in stratospheric particles agrees with earlier findings by Murphy et al. (1998; 2007; 2014).

Figure 3 shows the fractional abundance of the Fe~~iron~~ and Mg~~magnesium~~ particle type ~~binned~~ stratified by altitude, potential temperature, potential vorticity, and ozone, with bin sizes of ~~. For each bin (e.g., an altitude interval), the number of iron and magnesium type mass spectra recorded in this bin was divided by the total number of mass spectra recorded in this bin. The bin sizes are~~ 500 m for altitude, 4 K for potential temperature, 0.5 PVU (potential vorticity units, 1 PVU = $10^{-6}$ $m^2$ $s^{-1}$ K $kg^{-1}$) for potential vorticity, and 50 ppbv for ozone. In total, we detected 3140 particles of this type during ML-CIRRUS, 2412 during StratoClim 2016, 18688 during StratoClim 2017, 23138 during ND-MAX 2018, and 3310 during CAFE-Africa 2018 (see also Table 1). It has to be emphasized here that this fractional abundance refers to the total number of ~~analyze~~analysed particles by the ERICA and the ALABAMA. Both instruments use a 266 nm laser for ablation and ionization. Pure sulfuric

acid particles are not ablated and ionized at this wavelength, as was previously reported (Thomson et al., 1997; Murphy, 2007) and validated by laboratory measurements with the ERICA. Thus, the fraction of the ~~iron~~ Fe and ~~magnesium~~ 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.

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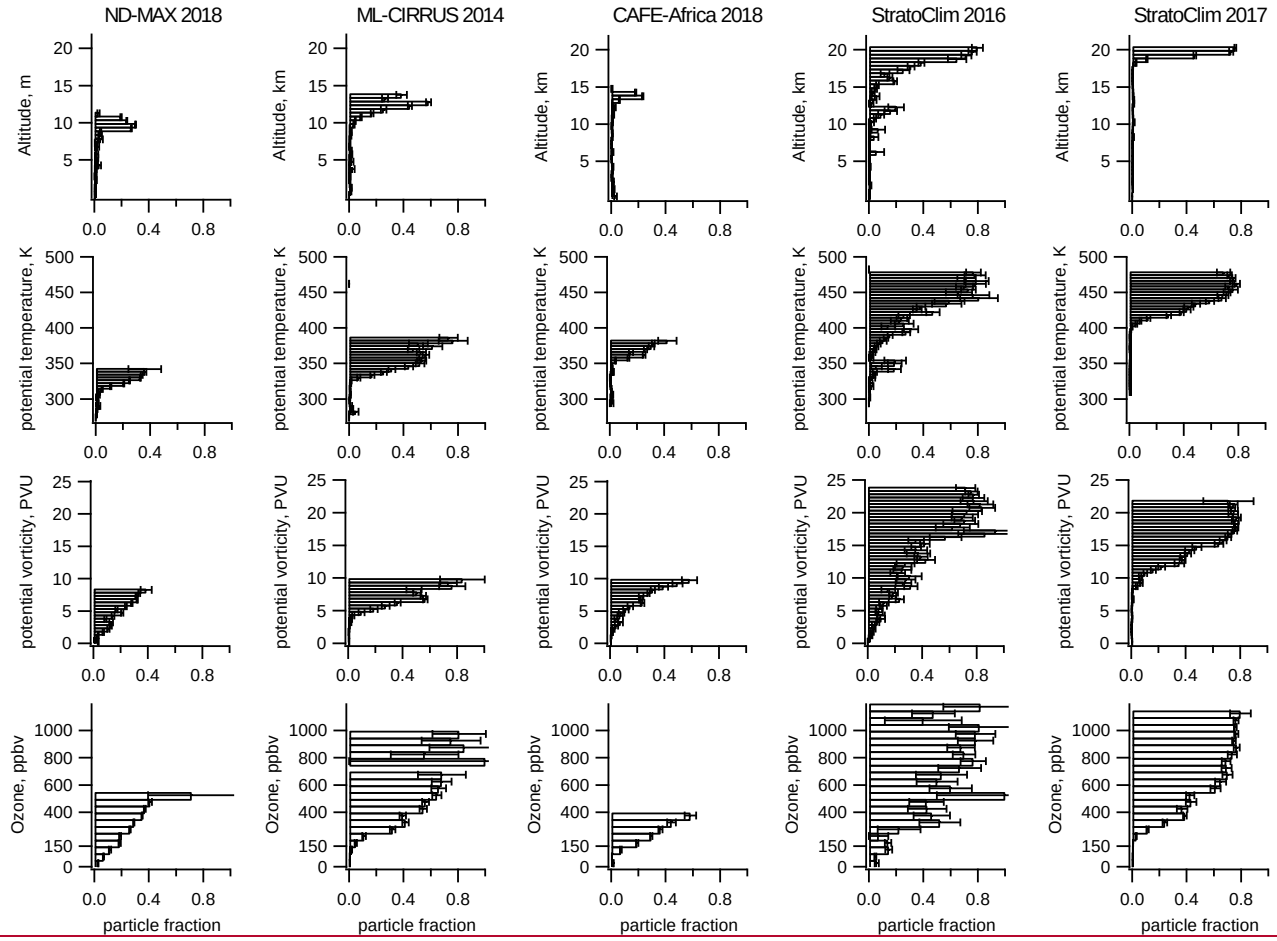

ND-MAX 2018
ML-CIRRUS 2014
CAFE-Africa 2018
StratoClim 2016
StratoClim 2017
Altitude, m
Altitude, km
potential temperature, K
potential vorticity, PVU
Ozone, ppbv
particle fraction

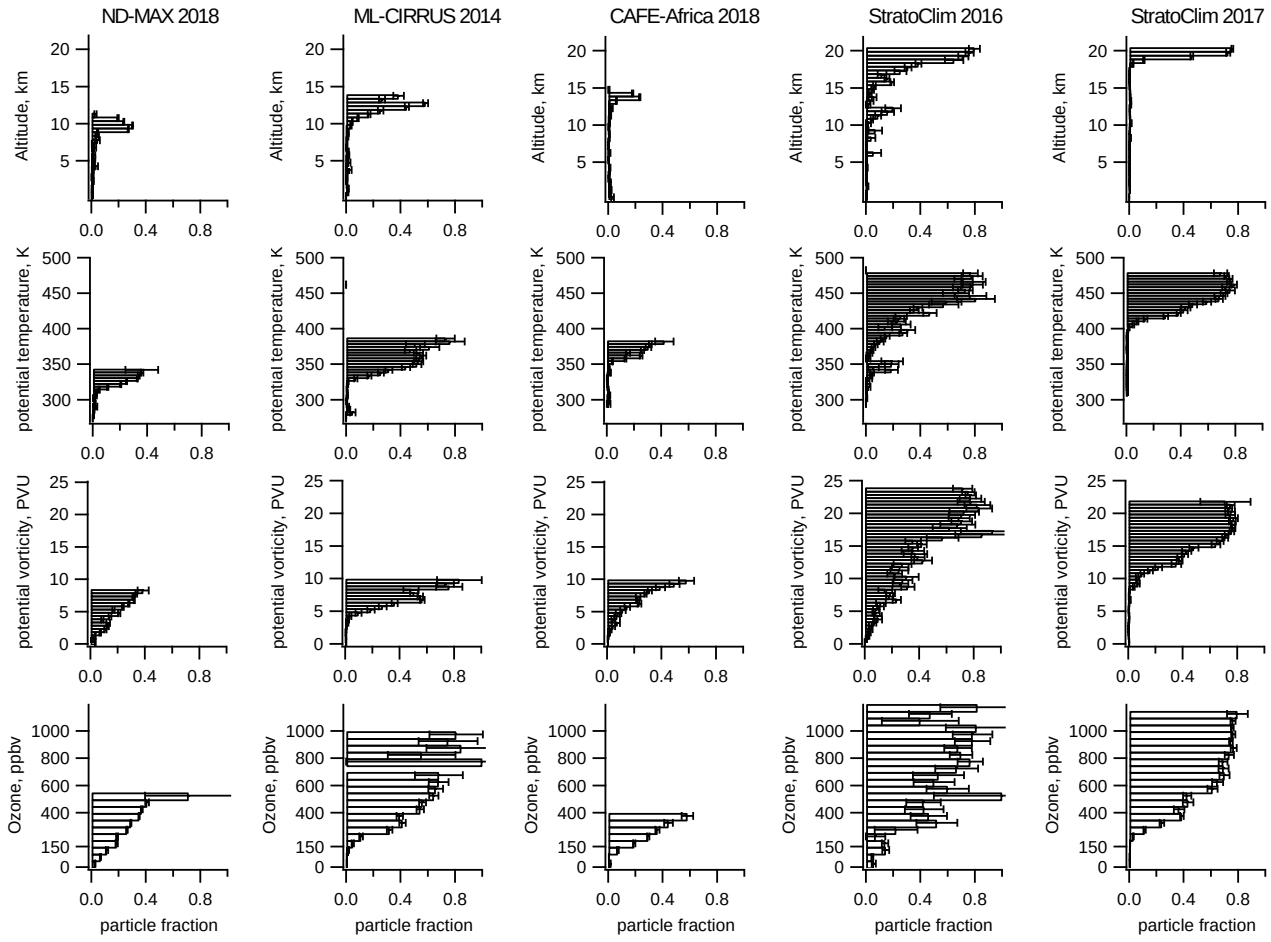


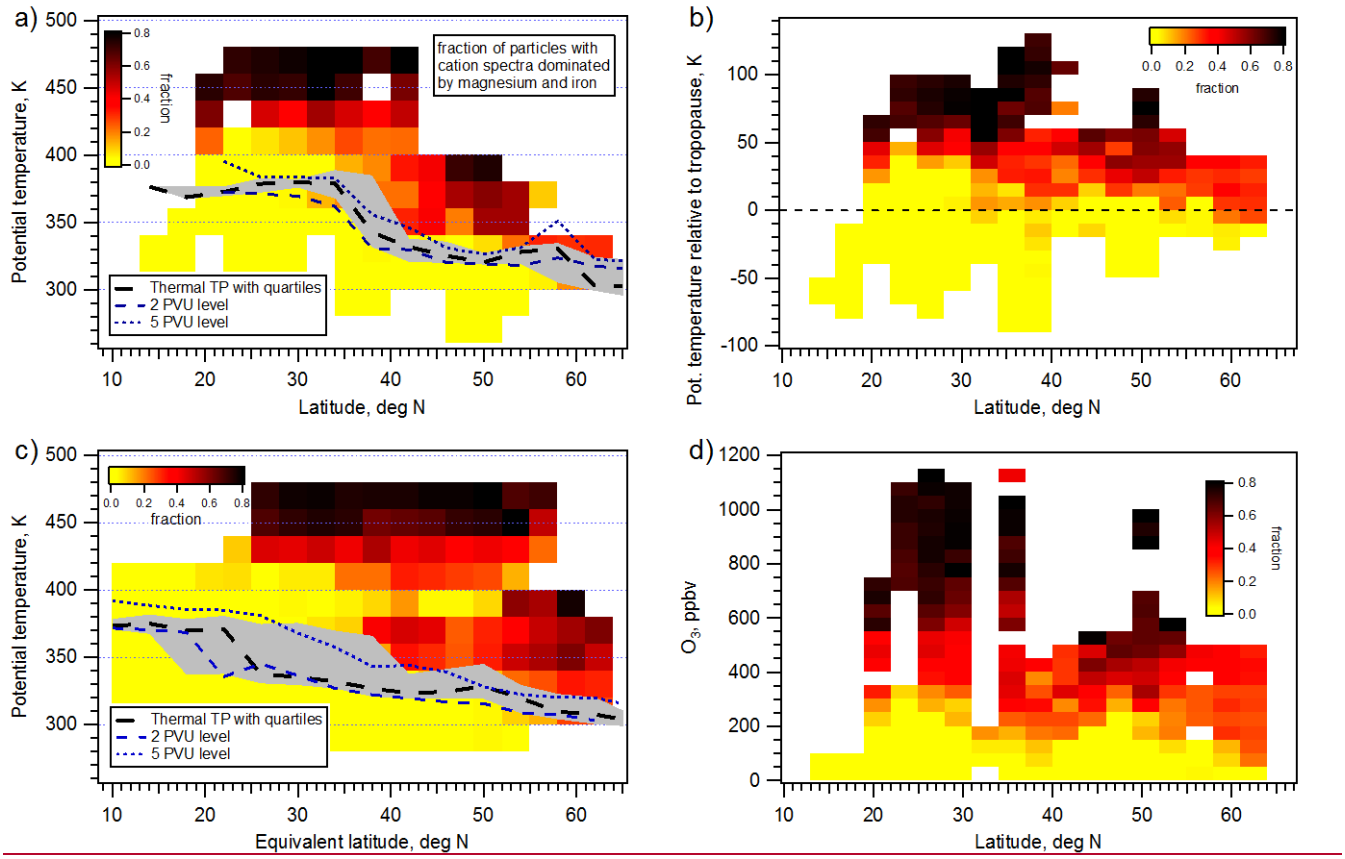

**Figure 3: Fractional abundance of particles with cation spectra dominated by magnesium and iron ions. Upper row: as function of geometric altitude; second row: as function of potential temperature; third row: as function of potential vorticity (PV); forth row: as function of ozone mixing ratio. The missions are not sorted in chronological order but from low potential temperature (leftmost column) to high potential temperature range (rightmost column). Error bars were calculated from Poisson counting statistics and propagation of uncertainty.**

During these five aircraft campaigns, the number of the Fe and Mg containing~~these~~ particles was largest at high altitudes (upper row in Fig. 3), where a fractional abundance of up to 0.8 was calculated. Similar values (up to 0.6) were reported by Murphy et al. (2014) for particles with the same ion signals in the mid-latitude stratosphere. The potential temperature and potential vorticity graphs (second and third rows) show that the high fractional abundance also corresponds to high values of potential temperature and potential vorticity, indicating that the measurements showing the high fractional abundance of this particle type were taken in the stratosphere.

The tropopause as the boundary between troposphere~~pause~~ and stratosphere is often defined via the temperature lapse rate (known as the thermal tropopause or cold-point tropopause). ~~, but~~The potential vorticity has been found to be a ~~good~~ better indicator, representing ~~for the~~a "dynamical tropopause" in the extratropics (Hoskins et al., 1985; Gettelman et al., 2011). The threshold value used to separate the stratosphere from the troposphere in the extratropics is typically 2 PVU (e.g., Holton et al., 1995), whereas this threshold value increases up to 5 PVU in the subtropics (Kunz et al., 2011). Here we find that the increase of this particle type fraction occurs at about 2 PVU during ND-MAX/ECLIF-2, StratoClim 2016 and CAFE-Africa, at 4 PVU during ML-CIRRUS, and at 8 PVU during StratoClim 2017. In general, PV is not well suited to define the tropopause level in the tropics and therefore, a potential temperature of 380 K is typically used instead of PV to define the tropical tropopause (Holton et al., 1995). Notably during the Stratoclim 2017 flights sampling ~~, which took place over~~above the AMA, the increase of the iron and magnesium containing particle fraction is found at 400 K, which is consistent with the high tropopause over the AMA. In the lowest row of Fig. 3, ozone is used as the vertical coordinate. Here, the increase of the particle fraction starts above an ozone mixing ratio of about 150 ppbv, indicating the chemical tropopause (Hoor et al., 2002; Zahn and Brenninkmeijer, 2003; Pan et al., 2004). The different tropopause altitudes observed during the individual missions are due to the fact that the height of the tropopause is a function of latitude. The tropical tropopause corresponds to an isentropic surface at a potential temperature level of about 380 K (Holton et al., 1995), corresponding to a geometric altitude of about 17 km (Fueglistaler et al., 2009). In the extratropics, the isentropes are crossing the dynamical tropopause that lies here between 2 and 5 PVU. At polar latitudes the tropopause height is typically around 8 km (Wilcox et al., 2012).

### 3.2 Latitudinal distribution

To combine all data from the five aircraft campaigns, we binned all particles (in total 338 354) by latitude and potential temperature, using 3° bins for latitude and 20 K bins for potential temperature ("theta"), thereby displaying the data in theta-latitude space. The same was done for the iron and magnesium-dominated particle type (in total 50688). Then we calculated the particle fraction of the magnesium-dominated particle type for each bin. Only bins containing more than 10 particles were considered. The result is shown in Fig. 4 a) (separated graphs for the individual missions are given in the supplement in Fig. S6). We also added to the figure ~~inserted~~ the thermal tropopause from the ECMWF data set, binned into 4-degree latitude bins. The median thermal tropopause is given by the thick dashed line and the 25% and 75% quartiles by the gray shaded area. Additionally, a range for the dynamical tropopause is shown, from 2~~2~~ PVU ~~and a~~to 5 PVU, ~~surface are shown~~ by the thin dashed lines~~, indicating the dynamical tropopause (2 PVU at mid-latitudes and 5 PVU in the subtropics)~~. For this, we took all potential temperatures where the potential vorticity ranged between 1.5 and 2.5 PVU (4.5 and 5.5 PVU, respectively) and binned these values into 4-degree latitude bins.


a)
Potential temperature, K
Latitude, deg N
fraction
fraction of particles with cation spectra dominated by magnesium and iron
Thermal TP with quartiles
2 PVU level
5 PVU level
b)
Pot. temperature relative to tropopause, K
Latitude, deg N
fraction
c)
Potential temperature, K
Equivalent latitude, deg N
fraction
Thermal TP with quartiles
2 PVU level
5 PVU level
d)
O3, ppbv
Latitude, deg N
fraction

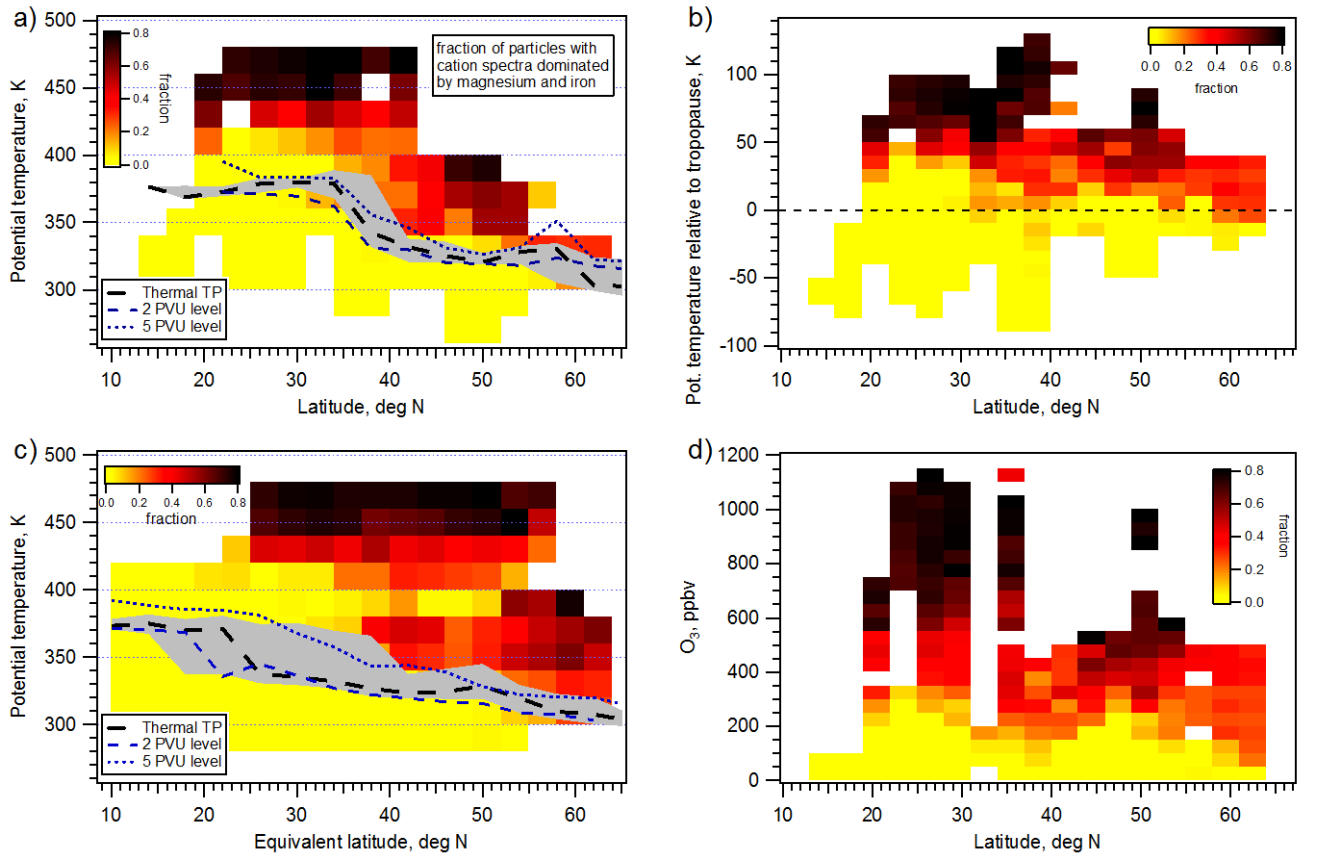



**Figure 4: Fractional abundance of particles with cation mass spectra dominated by magnesium and iron ions as a function of potential temperature and latitude (a), as a function of distance to the tropopause and latitude (b), as a function of potential temperature and equivalent latitude (c), and as a function of ozone and latitude (d). The data of all five UTLS aircraft missions have been merged for this figure (in total 338 354 ~~analyze~~analysed particles). Also shown in a) and c) is the median thermal tropopause (from ECMWF) along with interquartile ranges ~~s~~ (gray shading) and two dynamical tropopause levels (2 PVU and 5 PVU).**

The same procedure was used for the other panels in Fig. 4. In Fig. 4 b) the potential temperature relative to the tropopause is used as vertical coordinate with 10 K bins. Figure 4 c) uses equivalent latitude as horizontal coordinate. The equivalent latitude of an air parcel is calculated by transforming the contour having the same potential vorticity and potential temperature into a circle centered at the pole. The latitude enclosing this circle is then defined as the equivalent latitude. Since potential vorticity is conserved under adiabatic processes, equivalent latitude can be used to account for reversible adiabatic tracer transport by e.g. planetary waves (Hegglin et al., 2006; Hoor et al., 2010; Krause et al., 2018). Figure 4 d) uses ozone as vertical coordinate, with 50 ppb ozone bins.

In theta-~~geographical~~ latitude space (Fig. 4 a), the thermal tropopause reaches the 380 K level at 34°N and remains between 370 and 380 K south of 34°N. At mid-latitudes, the tropopause height decreases until it reaches 300 K at 60°N. In theta-

equivalent latitude space (Fig. 4c), the thermal tropopause shows more variation (larger interquartile range), especially between 20 and 40°N.

All sub-panels in Fig. 4 show that the fraction of the iron and magnesium-dominated particles increases in high and middle latitudes very close to the position of the tropopause, but not in the tropics. In theta-latitude space (Fig. 4 a), the particle fraction remains as low as in the troposphere between the tropopause (around 370 - 380 K) and 400 K at latitudes south of 30°N. Normalizing the potential temperature to the thermal tropopause (Figure 4b) confirms this observation. In theta-equivalent latitude space (Fig. 4 c), this effect is even more pronounced: South of 35°N equivalent latitude, the area between the tropopause and 420 K shows a very low fraction of the iron and magnesium-dominated particles. This corresponds to the PV profile of the StratoClim 2017 data from Figure 3, because the stratospheric tropical data in Fig. 4 are dominated by the StratoClim 2017 data set. In the AMA, which dominated the geographical region of StratoClim 2017 during the time of the campaign, the air masses are transported upwards between about 360 K and 460 K (Ploeger et al., 2017; Vogel et al., 2019). The observation that the fraction of the iron and magnesium-dominated particle type increases only above the extratropical tropopause layer or mixing layer (Hoor et al., 2002; Hoor et al., 2004; Pan et al., 2004), i.e. 30 K above the tropopause (Fig. 4 b), indicates that the source for this particle type must be above the tropopause, because otherwise, the upwelling air masses in the AMA would contain this particle type also at lower potential temperatures. In the stratosphere, the widespread occurrence of high fractions of this particle type over a broad range of equivalent latitudes above 440 K (Fig. 4 c) indicates that this particle type is very homogeneously distributed in the stratosphere. The large equivalent latitude range is consistent with potential transport between high and low latitudes. From Fig. 4 b) it can be seen that at above a distance of about 40 K to the tropopause the proportion of the iron and magnesium-dominated particles does not change substantially with latitude. In Fig. 4 d) similar behavior is observed for ozone levels larger than 300 ppbv.

### 3.3 Interpretation as meteoric particles

From the previous discussion we concluded that the source of this particle type is likely found above the tropopause. The capacity to record bipolar ion spectra of single particles allows us to show that each particle whose cation mass spectrum is dominated by Mg and Fe contains sulfuric acid but no other frequently observed anions like $NO^-$, $NO_2^-$, $CN^-$, or $CNO^-$. We therefore conclude that the particles we observe consist of meteoric material dissolved in sulfuric acid. This interpretation is fully consistent with the argumentation by Murphy et al. (1998) and Cziczo et al. (2001) who measured stratospheric particle composition using a similar laser ionization mass spectrometer (PALMS) on board the WB-57F high altitude research airplane between 5 and 19 km altitude. Additional PALMS measurements from other campaigns (Cziczo et al., 2001; Cziczo et al., 2004; Murphy et al., 2007; Murphy et al., 2014) as well as laboratory measurements with reference meteoric samples and artificial meteorite particles supported the conclusion that the stratospheric particles with mass spectra dominated by Mg and Fe consist of meteoric material dissolved in sulfuric acid (Cziczo et al., 2001). Our cation mass spectra (Fig. 2) show a very similar ion signature as the cation mass spectra from stratospheric particles, dissolved meteorites and artificial meteorite particles presented in Cziczo et al. (2001). The finding that Si is observed to a much lesser degree than expected from meteoric

composition (roughly equal amounts of Fe, Mg, and Si) was explained by Cziczo et al. (2001) and Murphy et al. (2014) by the low solubility of $SiO_2$ in $H_2SO_4$. Thus, Si is assumed to be present as a solid inclusion in the particles and is thereby less efficiently ionized compared to the other metals that are dissolved in $H_2SO_4$.

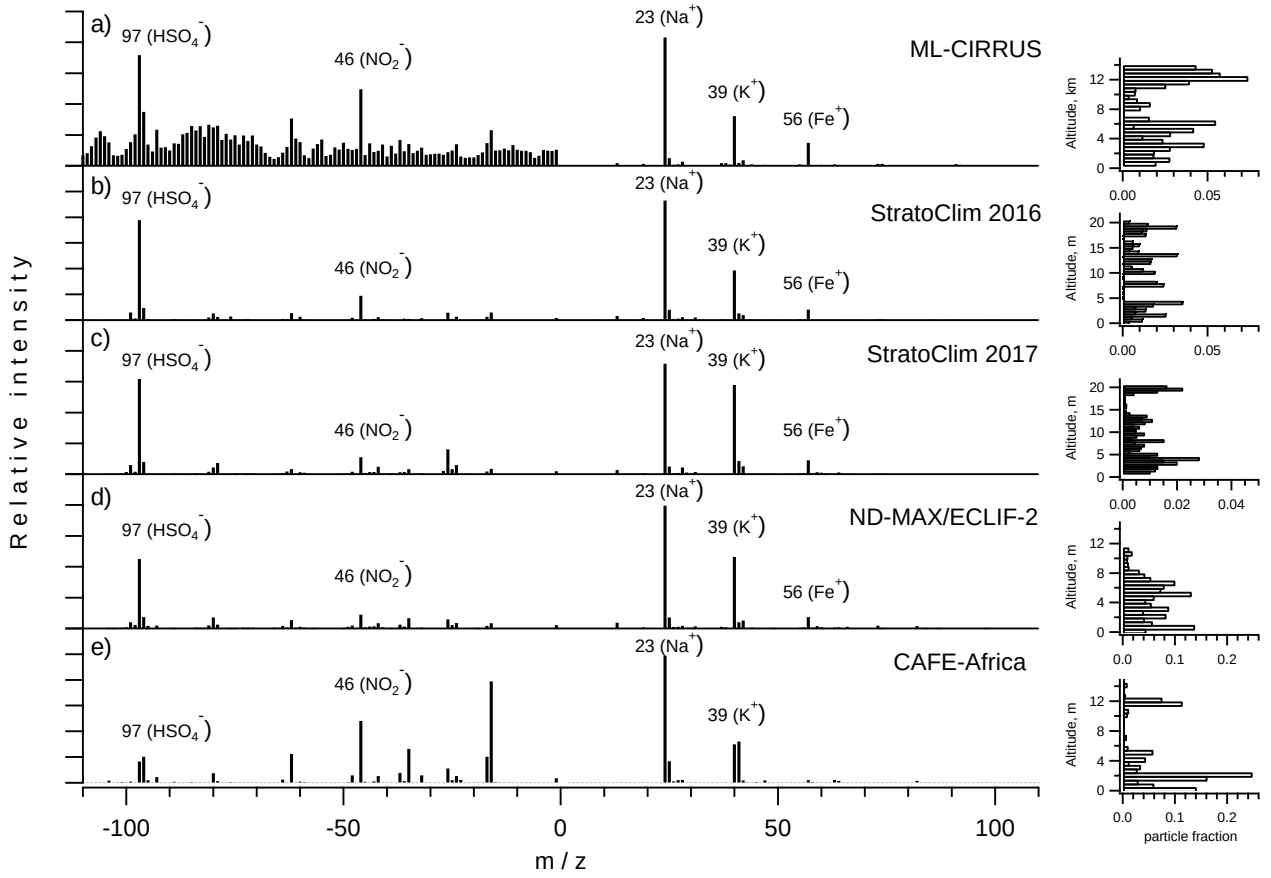



**Figure 5. Mean mass spectra and vertical profiles of a particle type containing Na, K, and Fe, with smaller amounts of Mg and Ca. This type, which was observed in all five high altitude aircraft missions, does not belong to the meteoric particles, although it was sometimes observed at higher altitudes. It can be interpreted as mineral dust, internally mixed with sea spray and secondary inorganic compounds (nitrate, sulfate). Note that during ML-CIRRUS the anion spectra were noisy due to problems with the high voltage supply of the ALABAMA.**

Other sources for this particle type, like aircraft or rocket exhaust, uplifting of particles (e.g. desert dust) from the Earth's surface and volcanic injection, can be ruled out. The majority of aircraft traffic does not occur at such high altitudes at which the meteoric particles were observed during the StratoClim campaigns. Rocket exhaust can be ruled out because the dominating metal in rocket exhaust particles is expected to be aluminium (Voigt et al., 2013). Single particle mass spectrometric measurements of rocket exhaust plumes showed ions of chlorine and oxygen, of metals like Al, Fe, Ca, Na, and K, but not of

magnesium (Cziczo et al., 2002). Furthermore, rocket exhaust plumes would hardly lead to the observed uniform and wide geographical distribution of the particle fraction. Volcanic aerosol particles have been measured in the tropopause region and lowermost stratosphere after eruptions of Kasatochi and Sarychev (Andersson et al., 2013). These data show that volcanic aerosol particles contain a larger weight percentage of carbonaceous material than of ash, which is not reflected in our measurements. Furthermore, volcanic ash particles indeed contain a number of elements that are abundant in meteorites, like Fe, Si, Ca, K, but additionally also elements that are characteristic for crustal material like titanium which was not observed in our mass spectra. As crustal material that can occur as particles in the troposphere (like soil dust or desert dust) contains the same elements like the stratospheric particles we observed (e.g., Na, Mg, Al, K, Fe), interferences with dust particles in the troposphere might be possible, although the ions $FeO^+$ and $FeOH^+$ (m/z 72, 73) have not been observed in single particle spectra of mineral dust (Gallavardin et al., 2008). In the tropical regions, uplifting of particles from the troposphere into the stratosphere occurs especially in the AMA (Randel et al., 2010; Pan et al., 2016; Yu et al., 2017) and might also carry dust particles into the stratosphere. However, to explain the stratospheric abundance-fraction of the observed Fe- and Mg-rich particle type, this particle type would need to be found already during the upward transport in the AMA, which is clearly not the case (Fig. 4). The mean mass spectra and the vertical profiles of another prominent particle type containing Fe, K, Na, as well as smaller signals of Mg and Ca, is shown in Fig. 5. This particle type was occasionally observed in the stratosphere (ML-CIRRUS, StratoClim 2017, CAFE-Africa), but in general occurred mainly in the troposphere. We interpret this particle type as an internal mixture of mineral dust, sea spray, sulfate, and nitrate, due to $Na^+$, $K^+$, and $Fe^+$ cations and chlorine ($^{35}Cl^-$, $^{37}Cl^-$), nitrate ($NO^-$, $NO_2^-$), and sulfate ($SO^-$, $SO_2^-$, $SO_3^-$, $HSO_4^-$) anions. It was therefore not included in the meteoric data set discussed in this paper. The reason why such particles were found in the stratosphere during ML-CIRRUS is presumably an outbreak of Saharan dust and its transport towards Europe during the time of the campaign (Weger et al., 2018). During StratoClim 2017 and CAFE-Africa, vertical uplifting of such particles of tropospheric origin into the stratosphere can most likely be explained by the Asian and African monsoon systems.

### 3.4 Size-resolved fraction of meteoric particles

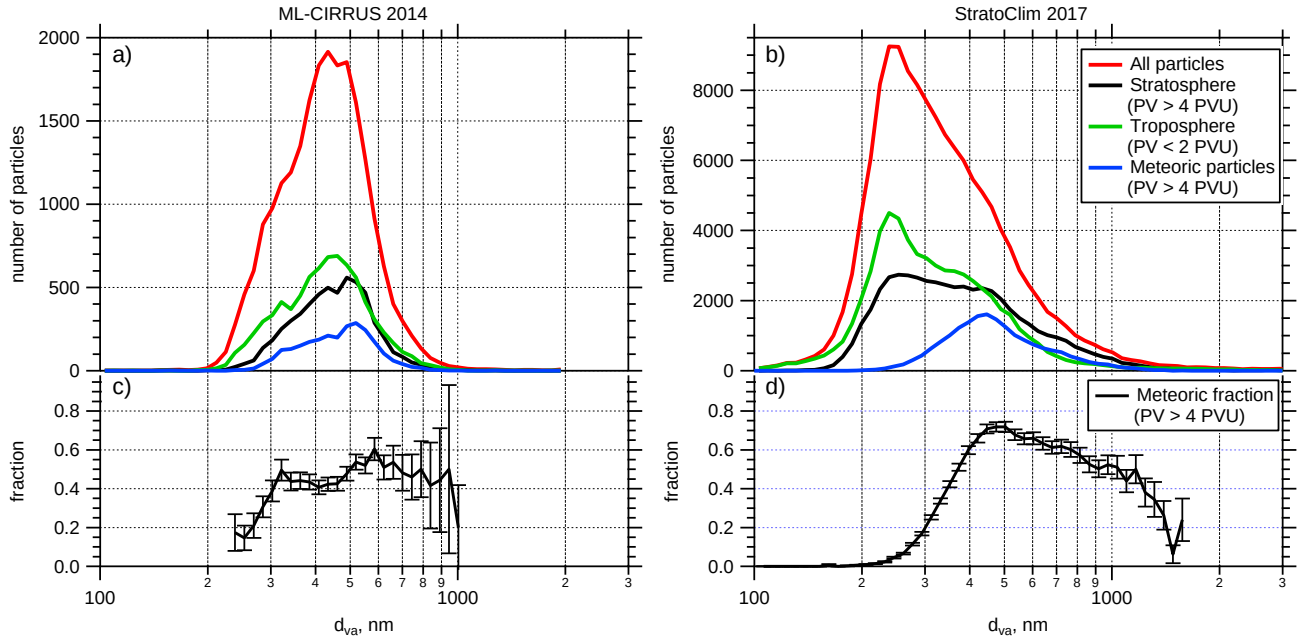



**Figure 6. Number of ~~analyze~~analysed single particles as a function of particle size (vacuum aerodynamic diameter, dva) measured during ML-CIRRUS 2014 (a, c) and StratoClim 2017 (b, d). Panels a) and b) show absolute number of counted particles per size bin; panels c) and d) show the fraction of particles with meteoric composition signature in the stratosphere (PV > 4 PVU). Error bars were calculated from Poisson counting statistics (number of particles per size bin) and were propagated for the particle fraction.**

Both particle mass spectrometers used here (ALABAMA and ERICA) determine the particle velocity in the vacuum chamber which by laboratory calibration can be converted into the vacuum aerodynamic diameter ($d_{va}$, DeCarlo et al. (2004)) of each individual particle. To obtain the size distributions shown in Fig. 6 a) and b), we used logarithmically equidistant size bins between 100 and 2000 nm (ML-CIRRUS) and 100 and 5000 nm (StratoClim). These size distributions represent the product of instrument efficiency (inlet transmission, particle detection and ablation rate) and the ambient particle size distribution. Differences between the measurements with the ALABAMA during ML-CIRRUS and the ERICA during StratoClim 2017 are therefore mainly due to the aforementioned differences in instrumental performance. The particle sizes were separated between tropospheric (PV < 2 PVU) and stratospheric (PV > 4 PVU) conditions.

In both data sets, the tropospheric particles (green lines) tend to be smaller than the stratospheric particles (black lines). Panels a) and b) in Fig. 6 also depict the size distribution of the meteoric particles, and panels c) and d) show the ratio between the meteoric particles (also selected for stratospheric conditions) and all stratospheric particles. It turns out that the fraction of meteoric particles is lowest in the smaller-particle size range for both campaigns: In the ML-CIRRUS data set, the fractional contribution increases from about 0.2 at 250 nm to about 0.5 at 300 nm and remains almost constant at 0.5 up to 1000 nm. The

StratoClim data set extends both to smaller and larger sizes and contains a larger number of particles. Here it can clearly be seen that the fraction of meteoric particles is zero at 200 nm, although stratospheric particles are detected even below 200 nm. The meteoric fraction rises to 0.7 at 450 nm and decreases above that size, down to 0.2 at about 1600 nm. Above that size, only one meteoric particle was detected, although in total 253 stratospheric particles were measured between 1600 and 4400 nm. Thus, the meteoric fraction appears to decrease down to zero above $d_{va} \approx 1600$ nm. This finding is similar to the data shown by Murphy et al. (2014) who found a maximum of meteoric particles at diameters of around 600 – 700 nm and a decrease down to zero above $d_{va} = 1$ µm. However, the fraction of meteoric particles below 600 nm is markedly higher in our data set compared to the study of (Murphy et al., 2014). In their data set, the fraction of meteoric particles decreases from 0.2 at 600 nm to zero at 500 nm, and no meteoric particles were detected below 500 nm diameter.

The observed size range of the meteoric particles between about 250 and 1500 nm indicates that their sedimentation may play an important role for the downward transport of meteoric material through the stratosphere (see Section 4). Once the meteoric aerosol particle material has reached altitude levels near the tropopause, its rapid removal out of the stratosphere due to cross-tropopause exchange and cloud formation processes is likely.

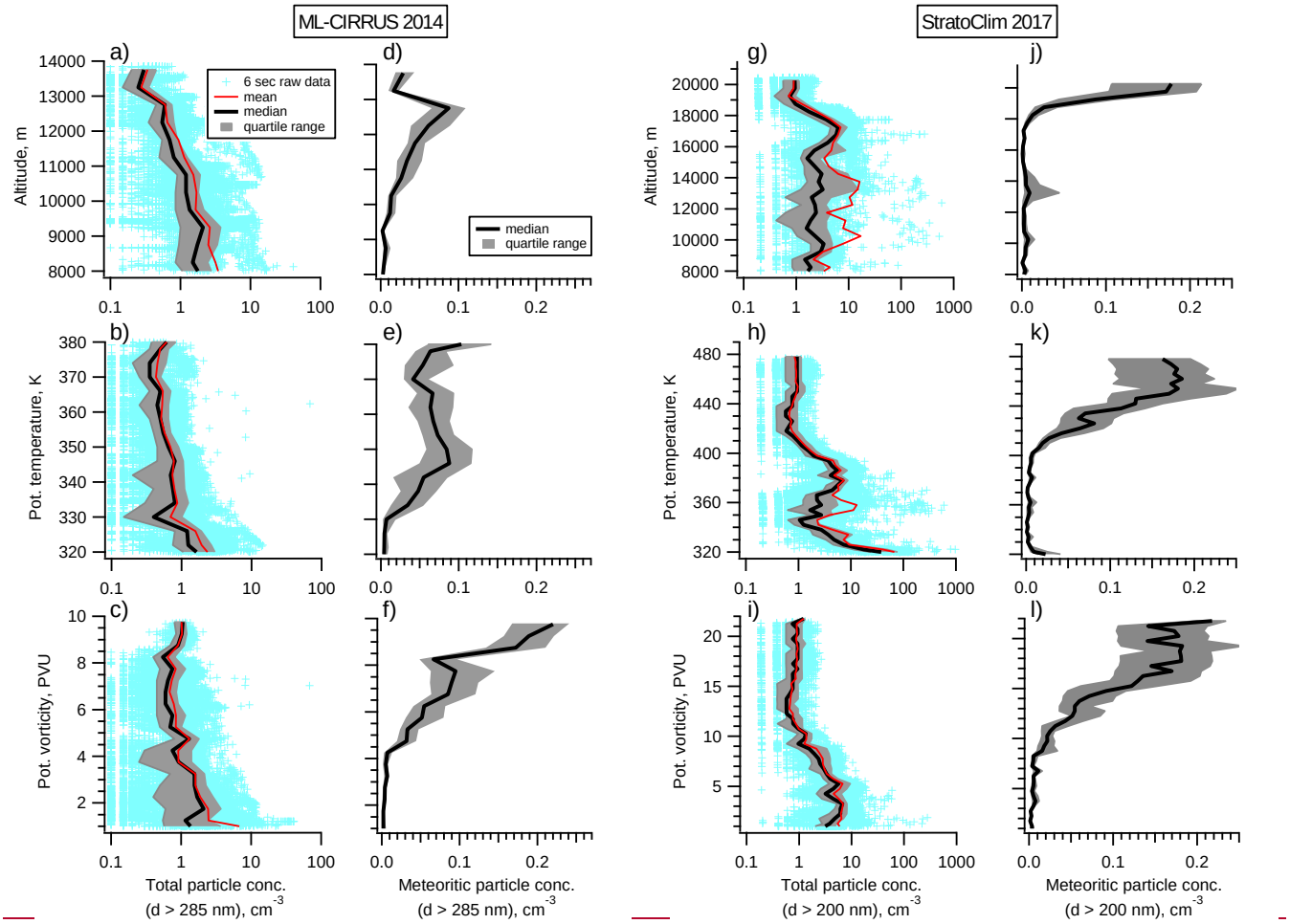
### 3.5 Particle number concentration

It is difficult to estimate accurately the~~an~~ absolute number concentration of particles containing meteoric material from the measured particle fraction with our~~a~~ 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 meteor~~it~~ic material will be overestimated due to the presence of~~if~~ pure sulfuric acid aerosol particles in the stratosphere~~existed in the air~~.

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, is 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 %.

~~The hit rate of the mass spectrometer, which is defined here as the number of acquired mass spectra per time unit divided by the number of laser shots per time unit, can be used to estimate the number of missed particles. Our data show that the hit rate~~

~~in the stratosphere is generally lower than in the lower troposphere. Two examples (for ML-CIRRUS and CAFE-Africa) are shown in Fig. S8 in the supplement. The maximum achieved hit rate in the troposphere was about 0.8 during CAFE-Africa, whereas the averaged hit rate in the stratosphere was about 0.2, thus, lower by a factor of 4. A similar decrease, albeit at lower absolute values of the hit rate, was observed during ML-CIRRUS. As a conservative approach, we assume here that the decrease of the hit rate in the stratosphere is only due to the abundance of pure sulfuric acid particles that are not ablated. Then~~ W~~w~~e ~~can~~ estimate the absolute number of meteoric particles ~~in two steps: First, by dividing the fraction measured by the mass spectrometer by a factor of 4 (to account for the hit rate decrease) and second,~~ by multiplying the meteoric particle fraction by the total particle number concentration measured using an independent absolute particle counting (and sizing) instrument. This process ~~e second step~~ is similar to previous approaches (Qin et al., 2006; Gunsch et al., 2018; Froyd et al., 2019).


ML-CIRRUS 2014
StratoClim 2017
a)
b)
c)
d)
e)
f)
g)
h)
i)
j)
k)
l)
6 sec raw data
mean
median
quartile range
Altitude, m
Pot. temperature, K
Pot. vorticity, PVU
Total particle conc.
(d > 285 nm), cm-3
Meteoritic particle conc.
(d > 285 nm), cm-3
Total particle conc.
(d > 200 nm), cm-3
Meteoritic particle conc.
(d > 200 nm), cm-3

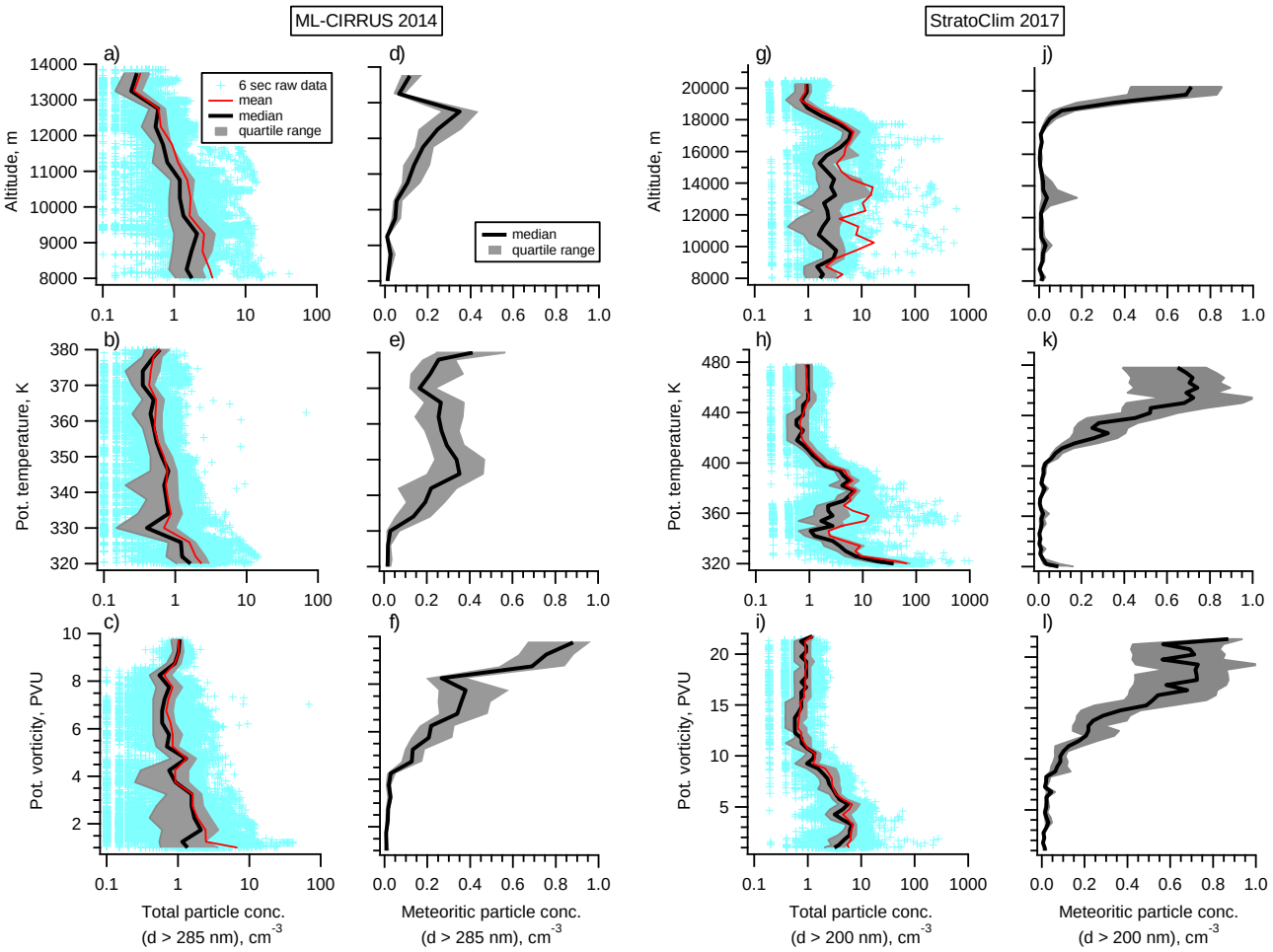



**Figure 7. Total particle number concentrations measured during ML-CIRRUS 2014 ($d_{ve}$ > 285 nm, a) to c)) and StratoClim 2017 ($d_{ve}$ > 200 nm, g) to i)) along with calculated number concentrations of particles containing meteoric material (ML-CIRRUS 2014: d) to f); StratoClim 2017: j) to l)). Data are shown for the upper troposphere and lower stratosphere (Altitude > 8 km, potential temperature > 320 K, PV > 1 PVU). Light blue markers: 6 second raw data; red line: mean; black line: median; grey area: quartiles (25% and 75%).**

For ML-CIRRUS, we used the optical particle spectrometer "Sky-OPC" (Grimm 1.129). The nominal lower cut-off diameter (manufacturer calibration~~ed~~ with PSL particles) is 250 nm. To account for the refractive index of stratospheric particles, we performed Mie calculations for refractive indices between 1.43 and 1.45 (Yue et al., 1994). This resulted in a lower cut-off diameter for stratospheric aerosol particles of 285 nm in diameter (supplement, Fig. S10~~4~~). The size distributions in Fig. 6 show that for ML-CIRRUS the meteoric fraction is approximately constant ~~between~~ above vacuum aerodynamic diameters greater than 300 nm. This value translates into a volume equivalent diameter ($d_{ve}$) of about 180 nm, assuming a density of the lower stratospheric particles of 1.7 g cm$^{-3}$ (Yue et al., 1994). We also note that the size distribution showed that 99.8 % of all particles counted by the OPC in the stratosphere are below 1000 nm. Thus, we can assume a constant fraction of meteoric

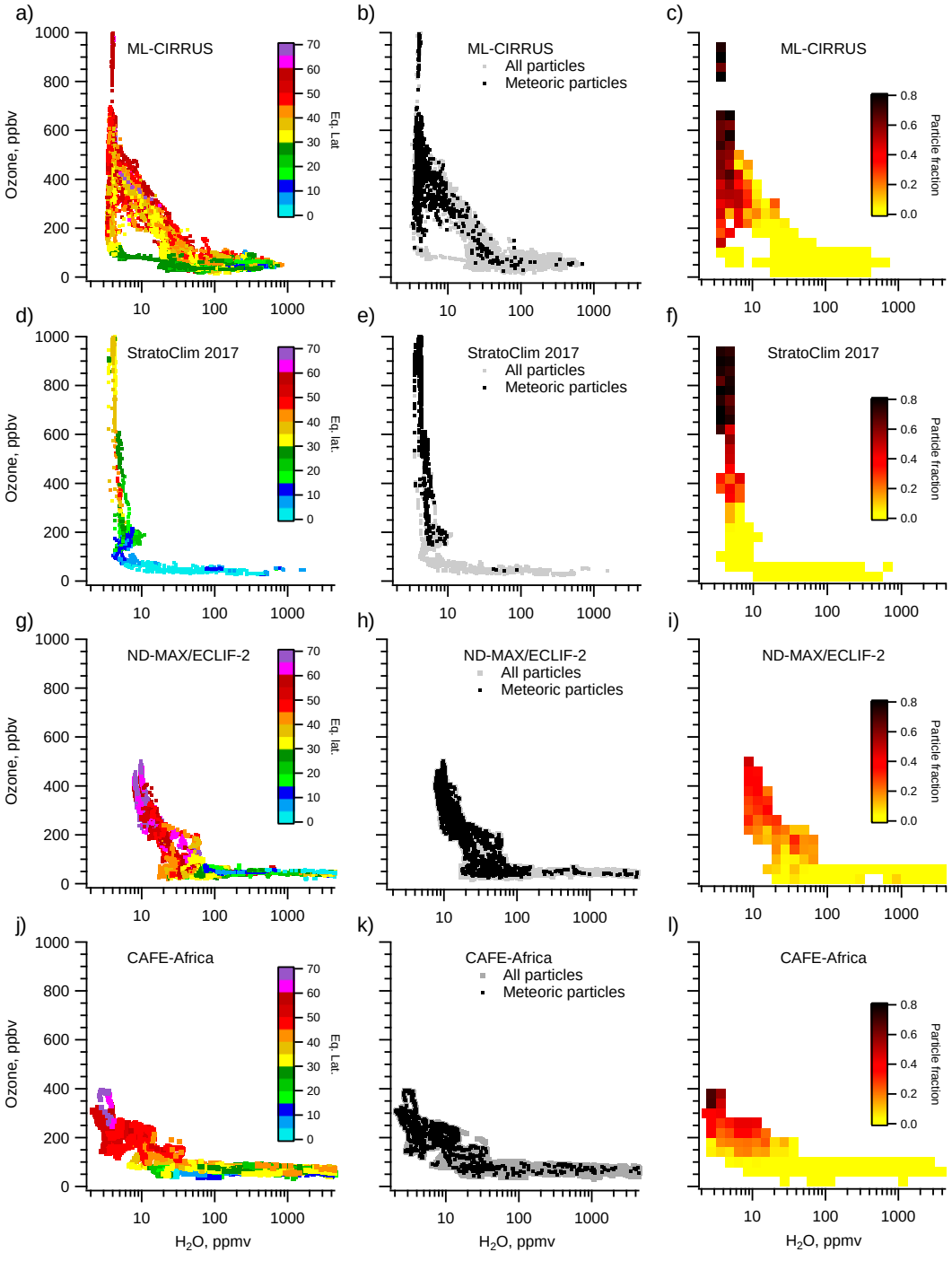
particles for the particles counted by the OPC and therefore multiplying the binned meteoric particle fraction ~~(divided by 4)~~ from Fig. 3 with the binned number concentration measured by the OPC should give an estimation of the absolute concentration of meteoric particles larger than 280 nm for the mid-latitude data set from ML-CIRRUS 2014.

For StratoClim 2017, we used data recorded by the UHSAS (DMT Inc.). According to the size distribution of meteoric particles in Fig. 6, the meteoric particles fraction reaches about 50% of its maximum fraction at 340 nm ($d_{va}$). This translates into a volume equivalent diameter ($d_{ve}$) of 200 nm (assuming the same density for stratospheric aerosol particles as above). Mie calculations using the refractive index range from 1.43 to 1.45 (Fig. S10~~1~~) yield that a lower size limit of 180 nm (PSL calibration) corresponds to a $d_{ve}$ of 200 nm for stratospheric aerosol particles. We therefore used the integrated particle number concentration between 180 nm and 1000 nm (PSL calibration), multiplied this with the fraction of meteoric particles from Fig. 3 ~~and divided by 4 to account for the hit rate~~. This procedure gives an estimation of the absolute concentration of meteoric particles larger than 200 nm for the tropical data set from StratoClim 2017.

Figure 7 shows the total particle concentrations for the two missions named above as a function of altitude, potential temperature and potential vorticity for the upper troposphere and lower stratosphere. The 6 second raw data are shown along with binned mean, median, and quartiles. The number concentrations of about 1 $cm^{-3}$ at 20 km for particles greater than 200 nm are in agreement with previous balloon-borne stratospheric measurements (Deshler et al., 2003; Renard et al., 2020). ~~-~~The calculated meteoric particle concentrations are shown as binned median values with quartiles. The highest absolute number concentrations of meteoric particle range around 0.~~2~~8 $cm^{-3}$ (referring to ambient pressure and temperature). During StratoClim 2017 these values are reached above 20 km, potential temperature = 50 K, and 17 PVU. During ML-CIRRUS, values of 0.~~2~~8 ~~-~~$cm^{-3}$ are only reached at PV > 9 PVU, whereas in altitude and potential temperature coordinates the concentrations reach only 0.~~4~~1 $cm^{-3}$. Nevertheless, the absolute range of meteoric particle concentration is very similar for both data sets, although the calculation of the meteoric particle concentration relies on different size ranges of the optical instruments~~, and~~ is based on several assumptions and suffers from various uncertainties, as detailed above.

### 3.6 Transport mechanism for cross-tropopause exchange

To investigate the downward transport of meteoric particles through the tropopause into the troposphere, we use the tracer-tracer correlation of ozone as a stratospheric tracer and water vapor as a tropospheric tracer. Tracer-tracer scatter plots have been widely used to identify mixing between troposphere and stratosphere (Fischer et al., 2000; Hoor et al., 2002; Pan et al., 2004; Marcy et al., 2007; Gettelman et al., 2011; Krause et al., 2018). In such scatter plots irreversible tracer exchange shows up as lines connecting the respective mixing ratios of the initial unmixed reservoir air parcels and are termed mixing lines (Hoor et al., 2002). Occurrence of mixing lines is a clear indication for mixing between stratospheric and tropospheric air. It is more common to use carbon monoxide as a tropospheric tracer, but because it was not measured during ML-CIRRUS, we use water vapor for which the applicability to serve as a tropospheric tracer in tracer-tracer correlations has been shown by Gettelman et al. (2011), Pan et al. (2014) and Heller et al. (2017). High ozone values indicate stratospheric air (vertical branch), high $H_2O$ values tropospheric air (horizontal branch).


a)
ML-CIRRUS
Ozone, ppbv
Eq. Lat
b)
ML-CIRRUS
All particles
Meteoric particles
c)
ML-CIRRUS
Particle fraction
d)
StratoClim 2017
Ozone, ppbv
Eq. lat.
e)
StratoClim 2017
All particles
Meteoric particles
f)
StratoClim 2017
Particle fraction
g)
ND-MAX/ECLIF-2
Ozone, ppbv
Eq. lat.
h)
ND-MAX/ECLIF-2
All particles
Meteoric particles
i)
ND-MAX/ECLIF-2
Particle fraction
j)
CAFE-Africa
Ozone, ppbv
Eq. Lat.
k)
CAFE-Africa
All particles
Meteoric particles
l)
CAFE-Africa
Particle fraction
H2O, ppmv
H2O, ppmv
H2O, ppmv

610

**Figure 8. Ozone mixing ratio as stratospheric tracer versus water vapor mixing ratio as tropospheric tracer color-coded with equivalent latitude, for ML-CIRRUS (a), StratoClim 2017 (d), ND-MAX/ECLIF-2 (g), and CAFE-Africa (j). Panels b), e), h), and k) show $O_3$ and $H_2O$ for the times when particle mass spectra were recorded (gray: all particles, black: meteoric particles). Panels c), f), i), and l) show the proportion of meteoric particles within a $O_3$-$H_2O$ grid.**

Figure 8 shows the tracer-tracer correlations between ozone and $H_2O$ for ML-CIRRUS, StratoClim 2017, ND-MAX/ECLIF-2, and CAFE-Africa. The data coverage of $O_3$ and $H_2O$ during StratoClim 2016 was not sufficient (see supplement, Fig S7~~10~~). The left panels show all data from the trace gas measurements, color coded by equivalent latitude.

The mid-latitude data from ML-CIRRUS (Fig. 8 a) show a clear separation between air masses of mid latitude and tropical origin. The mixing lines, indicating irreversible mixing between the troposphere and the stratosphere have equivalent latitudes > 30°N, whereas the green colored data points that correspond to tropical air masses (equivalent latitude < 30°N) do not show such mixing. Fig. 8 b) shows the $H_2O$ and $O_3$ data for all sampled particles (gray) and for all meteoric particles (black). As expected, the density of black data points is highest in the stratospheric branch of the tracer-tracer correlation. Figure 8 c) shows the fraction of meteoric particles within an ozone-water-vapor grid. Mixing between extratropical stratospheric and tropospheric air is indicated by mixing lines with equivalent latitudes > 30°N, connecting regions of elevated extratropical and low stratospheric $H_2O$ values. Isentropic mixing between dry air which passed the Lagrangian cold point (and therefore exhibits $H_2O$ mixing ratios < 6 ppmv) and higher latitudes is indicated by the vertical branch starting at $O_3$ mixing ratios < 150 ppbv, connecting the dry upper tropical troposphere with the stratosphere.

In the StratoClim 2017 data set (Fig. 8 ~~c~~d), ~~d~~e), ~~e~~f)) no mixing lines were observed. Only very few meteoric particles are observed in the tropospheric branch of the $O_3$-$H_2O$ plot (below 100 ppbv $O_3$), showing that downward mixing of meteoric particles from the stratosphere does not occur in the upwelling tropical air masses of the AMA (see also Fig. 4).

The data sets of ND-MAX/ECLIF-2 (Fig. 8 ~~f~~g), ~~g~~h), ~~h~~i)) and CAFE-Africa (Fig. 8 ~~i~~j), ~~j~~k), ~~k~~l)) appear similar in this tracer-tracer correlation, although the geographic latitudes and seasons of the two campaigns were very different. In both missions, highest observed $O_3$ values are 400 – 500 ppbv, and the equivalent latitudes reach up to 60 – 70°N in the stratosphere. Both data sets show a high degree of stratosphere-troposphere mixing, as can been seen from the higher $H_2O$ mixing ratios at $O_3$ levels between 100 and 200 ppbv, corresponding to the mixing lines observed during ML-CIRRUS. Along these mixing lines, meteoric particles are frequently observed, even at tropospheric altitudes where water vapor mixing ratios of > 1000 ppmv are reached.

### 3.7 Detection of particles containing meteoric material at low altitudes

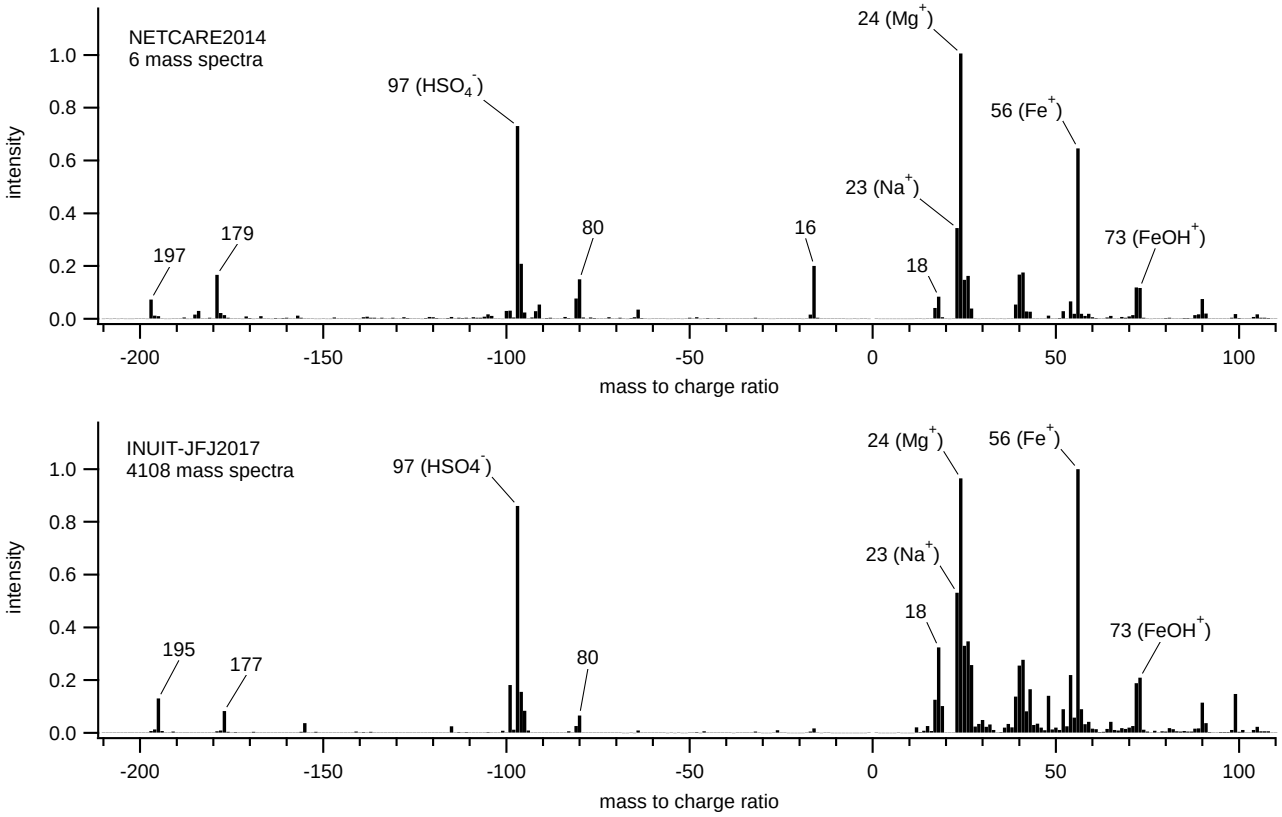



**Figure 9: Mass spectra from low altitudes (NETCARE, Canadian Arctic, summer 2014, up to 3 km; Jungfraujoch, Switzerland, winter 2017, 3600 m) showing meteoric signatures.**

Figure 8 showed that particles containing meteoric material are transported downwards through stratosphere-troposphere exchange and are therefore also present in the troposphere, albeit at low concentrations and low number fractions. We used two data sets from lower altitudes to estimate the occurrence of this particle type in the middle and lower troposphere. These are the abovementioned NETCARE data set (Canadian Arctic, summer 2014) that contains aircraft-based ALABAMA data up to 3 km altitude (Köllner et al., 2017) and a mountain-top data set from the Jungfraujoch station at 3600 m altitude in winter 2017. In both data sets the relative number of meteoric particles was very low (0.0025 – 1%), such that an automated cluster algorithm would not find this particle type unless the prescribed number of cluster would be set to very high values. Thus, we

used the most prominent mass spectral features from this particle type as observed in the stratosphere (Fig. 2) and scanned the two low-altitude data sets for these marker peaks. The criteria included the presence of $^{24}Mg$, $^{25}Mg$, $^{26}Mg$, $^{54}Fe$, $^{56}Fe$, the absence of Cl (to exclude sea spray) and signal intensity of m/z 39 smaller than that of m/z 41. The latter was introduced to minimize the influence of potassium from other sources, especially dust. By varying these search criteria, different numbers of mass spectra with similar average mass spectra were obtained, such that the absolute amount of meteoric particle at low altitude is highly uncertain. Figure 9 shows the averaged mass spectra matching the criteria given above. The spectra correspond very well to the spectra sampled in the stratosphere (Fig. 2). A higher contribution of m/z 18 ($NH_4^+$), especially in the Jungfraujoch spectra indicates a higher degree of neutralization of the sulfuric acid by ammonia in the troposphere than in the stratosphere. During NETCARE, six out of about 10000 particle mass spectra matched the criteria. By changing the criterion for potassium to an absolute upper intensity threshold, the number of spectra was reduced to three. Thus, the fraction of meteoric particles found in the summer Arctic lower troposphere can be estimated to be around 0.025 – 0.05%. In the free tropospheric data set obtained in winter at the Jungfraujoch, about 4100 spectra (out of more than 765000) matched the criteria, corresponding to 0.5%. Also here, by varying the criteria the percentage varies between 0.2 and 1 %. This range is clearly larger than that in the Arctic summer, but it has to be kept in mind that the Jungfraujoch data set was obtained at 3600 m altitude, whereas the measurements during NETCARE only reached up to 3000 m. In winter the Jungfraujoch station is mainly located in the free troposphere (over 60% of the time, see Bukowiecki et al. (2016)), such that the influence of boundary layer particles is low. In contrast, the aerosol particles in the summer Arctic during NETCARE were to a large degree influenced by particles from marine biogenic origin (Köllner et al., 2017). Backtrajectory calculations for the Jungfraujoch data set showed that the fraction of detected meteoric particles was higher during times when the air masses experienced higher altitudes and higher latitudes during the 5 days before the measurements (supplement, Section ~~9~~ 8 and Fig. ~~S12~~S11). Additionally, the fraction of meteoric particles followed the time trend of the ozone mixing ratio (Fig. ~~S12~~S11), confirming the stratospheric origin. Overall, this shows that the meteoric material immersed in stratospheric sulfuric acid aerosol particles reaches the lower troposphere from where it will be removed by wet removal (rain-out, wash-out), thereby finally reaching the Earth's surface. This is confirmed by a number of studies that reported the detection of meteoric material in ice cores samples from Greenland (Gabrielli et al., 2004; Lanci et al., 2012) and Antarctica (Gabrielli et al., 2006).

## 4 Discussion and conclusion

In this study we present stratospheric single particle mass spectrometer data from five aircraft-based campaigns, covering a wide range of northern hemispheric latitudes (15°N – 68°N) and seasons (winter, spring, summer). In all data sets a distinct particle type characterized by iron and magnesium was observed in the stratosphere. The observed distribution as function of potential temperature and potential vorticity suggests that the source of this particle type is above the tropopause. Consistent with ~~From~~ previous stratospheric aerosol composition measurement data (Mossop, 1965; Murphy et al., 1998; Cziczo et al., 2001; Murphy et al., 2014), meteoric composition data (Lodders and Fegley Jr., 1998; Rapp et al., 2012; Plane et al., 2015),

and theory of meteoric ablation and fragmentation (Plane, 2003; Carrillo-Sánchez et al., 2016; Subasinghe et al., 2016), it ~~i~~was concluded that this particle type represents meteoric material partially dissolved within sulfuric acid solution droplets.

Downward transport of meteoric smoke~~MSP~~ particles from the mesosphere into the lowermost stratosphere occurs efficiently in the polar vortex (Curtius et al., 2005; Weigel et al., 2014). These papers show that high altitude aircraft measurements demonstrate there is a higher proportion of refractory particles (60-70%) within the wintertime polar vortex, and one would therefore expect to find a higher abundance of meteoric particles in the lower stratosphere at high latitudes during late winter and early spring than at lower extra-tropical latitudes and than in other seasons. This expectation would largely agree with (1) the results by Dhomse et al. (2013), who predicted a more effective (by a factor of ~15) deposition of meteoric ablation material over Greenland than in Antarctica, and (2) the works of Bardeen et al. (2008) and Brooke et al. (2017), according to which the meteoric ablation material most effectively subsides to stratospheric altitudes within the polar winter vortex. This is not confirmed by our observations: Although two mid-latitude campaigns (ML-CIRRUS and ND-MAX/ECLIF-2) were conducted between January and April, we observe the same fraction of meteoric particles at the same ozone levels in the lower stratosphere (Fig. 3) during all campaigns, regardless of latitude and season.

Satellite observations with the CALIOP lidar instrument have shown that the lower edge of the stratospheric aerosol layer lies between 450 and 500 K potential temperature for latitudes between 20 and 50°N (Vernier et al., 2009). It is important to note however, that Aitken-sized particles (a few tens of nanometers, e.g. Cadle and Kiang, 1977) are present throughout the upper troposphere and lower stratosphere, ~~so~~ with this altitude range from the CALIOP measurements ~~refers~~ referring only to the optically-interacting aerosol population. Thus, we conclude that the particles containing meteoric material that we observed in the lower stratosphere (with diameters greater than 200 nm) originate from the stratospheric aerosol layer. Our measurements between 20 and 40°N reach up to the lower edge of the stratospheric aerosol layer (see Fig. 4), whereas at higher latitudes and lower altitudes we assume that we observed particles that settled gravitationally from the stratospheric aerosol layer. This in turn means that the stratospheric aerosol layer particles contain meteoric material at all latitudes. We therefore conclude that meteoric material is indeed carried within the winter polar vortex from the mesosphere to the stratosphere, but that isentropic mixing above 440 K potential temperature (see Fig. 4) distributes the meteoric material over all latitudes. Isentropic mixing in the stratosphere within the extratropics, but also between the tropics and extratropics, has been described previously (Neu and Plumb, 1999; Garny et al., 2014). From our data we can infer that at the lower edge of the stratospheric aerosol layer (above 440 K potential temperature) the meteoric material is equally distributed throughout the latitude range of about 20 to 60°N.

Our observations of particles with signatures of meteoric material do not clearly indicate the formation history, i.e. whether the material originates from meteoric disintegration by ablation (MSP), fragmentation (MF) or from interplanetary dust particles (IDP), since the material is at least partially dissolved in sulphuric acid.~~Our observations do not give a clear indication whether the detected particles containing meteoric material originate from meteor smoke particles (MSP) dissolved in stratospheric aerosol layer particles or from meteoric fragments (MF) or unablated interplanetary dust particles (IDP) that are coated by sulfuric acid. However, the high $H_2SO_4$ content of all detected meteoric particles and the uniform mass spectra suggest that MSP dissolved in sulfuric acid are the most likely particle source.~~

We calculated the terminal settling velocity for particles of different sizes and densities (pure $H_2SO_4$, $\rho$ = 1.83 g $cm^{-3}$, and pure olivine as a surrogate for meteoric composition, $\rho$ = 3.30 g $cm^{-3}$) as a function of altitude (for details see supplement, Section S5). Between 16 and 18 km, the settling velocity ranges between 1 and 12 m/day for particles between 100 and 500 nm having the densities given above. In the AMA, air masses are transported upwards between about 360 K and 460 K with about 1.5 K/day (Ploeger et al., 2017; Vogel et al., 2019), corresponding to about 35 – 40 m / day. This is larger than the above calculated range, thus sedimentation is not fast enough to overcome the Asian monsoon upward motion. This explains that in the tropics we observe the increased fraction of particle containing meteoric material only 30 K above the thermal tropopause (see Fig. 4) whereas in the extratropics, where little upward motion occurs, we see these particles directly above the tropopause. Our data further show that all meteoric particles contained $H_2SO_4$, but no other anions like nitrate or organic material. Thus, from our simultaneous cation and anion measurements we can confirm previous assumptions that Mg and Fe are dissolved in $H_2SO_4$ (Murphy et al., 1998; Cziczo et al., 2001; Murphy et al., 2014). This suggests that these particles act similar as pure $H_2SO_4$ droplets in the UT with respect to cirrus formation, but it is conceivable that dissolved meteoric material alters the viscosity of $H_2SO_4/H_2O$ droplets which was found to increase at low temperatures (Williams and Long, 1995). With respect to PSC formation in the ~~and also in the~~ polar stratosphere, ~~with respect to PSC formation.~~the works by James et al. (2018) and Tritscher et al. (2019) showed that "foreign nuclei" are needed to be included in their simulations to reproduce PSC observations. The finding that meteoric material is mainly present as dilute solution in $H_2SO_4/H_2O$ droplets needs to be included in future simulations.

~~-~~In general, the meteoric~~is~~ particle type represents a good tracer for stratospheric aerosol particles. Downward transport along mixing lines at mid-latitudes was clearly identified, but only for equivalent latitudes above 30°N. In data sets acquired in the lower troposphere the meteoric composition-signature particles were detected as well, albeit only in very minor fractional abundance. Their size and composition (larger than 200 nm, composed mainly of $H_2SO_4$, most likely neutralized by ammonia in the troposphere) makes them ideal CCN, such that they will be efficiently removed from the atmosphere by nucleation scavenging and wet removal and the meteoric material is by these processes transported to the Earth's surface. We re-iterate that our findings of relatively invariant meteoric-particle-fraction are for particles larger than 200 nm. The data from the Arctic campaigns (Curtius et al., 2005; Weigel et al., 2014) are for particles larger than 10 nm, including also stratospheric Aitken-sized particles, which will have a different abundance-fraction of particles containing meteoric material. The analysis presented here, combining data obtained with two different laser ablation mass spectrometers during five aircraft missions conducted at different seasons, latitudes, and altitudes, as well as two low altitude data sets, has confirmed the widespread occurrence of meteoric material in stratospheric aerosol particles. Using the particles containing meteoric material as a tracer for stratospheric transport, our observations confirm the upward motion of air masses over the Asian monsoon anticyclone and the associated transport into the stratosphere, the exchange between stratosphere and troposphere in the extratropics as well as the efficient isentropic mixing between high and low latitudes above 440 K potential temperature.

## Data availability

The data shown in this study are available at Edmond – the Open Access Data Repository of the Max Planck Society, under the following permanent link: https://dx.doi.org/10.17617/3.38

## Author contribution

JS evaluated the data, compiled the figures, and drafted the manuscript with contributions by RW. PHOO contributed significantly to the discussion on cross-tropopause mixing. Single particle mass spectrometer data were provided by OA, AH, AD, SM, SB (ERICA) and JS, TK, FK, OE, HCC (ALABAMA). JUG provided meteorological re-analyses. CM provided UHSAS data. AZ, FO, HS, MS, FR, AU provided ozone data. MK, CR, MZ, GSD, JPD, JBN provided water vapor data. PHOP provided information on meteoric composition. The manuscript was critically reviewed by RW, SB, OA, FK, HCC, AH, OE, PHOP, PHOO, MK, CR, JUG.

## Competing interests

The authors declare no competing interests

## Acknowledgements

This work was funded by the DFG Priority Program SPP 1294, grant SCHN1138/1-2 (ML-CIRRUS), by the European Research Council (ERC), EU FP7/2007–2013, Projects No. 603557 (StratoClim), and No. 321040 (EXCATRO). StratoClim was also supported by BMBF under the joint ROMIC-project SPITFIRE (01LG1205A). CAFE-Africa was funded by the Max Planck Society. ND-MAX was funded by the NASA Advanced Air Vehicles Program (program manager J. Dryer). Funding for NETCARE was provided by the Natural Sciences and Engineering Research Council of Canada through the NETCARE project of the Climate Change and Atmospheric Research Program. The measurements at Jungfraujoch were funded by the DFG grant SCHN1138/2-2 (FOR1525 "INUIT") and by the EU Horizon 2020 research and innovation programme, grant No. 654109 (ACTRIS-2). We also acknowledge that the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG), 3012 Bern, Switzerland, made it possible for us to carry out our experiment at the High Altitude Research Station at Jungfraujoch. The Swiss Federal Laboratories for Materials Science and Technology (EMPA) is acknowledged for providing ozone data for the Jungfraujoch through the EBAS/EMEP data base. We acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http://www.ready.noaa.gov) used in this publication. We thank the technical and scientific coordinators of the aircraft campaigns: C. Voigt, A. Minikin, U. Schumann (ML-CIRRUS), J. Lelieveld, H. Fischer, J. Williams, M. Dorf (CAFE-Africa), B. Anderson, T. Moes (ND-MAX/ECLIF-2), F. Stroh, M. Rex, F. Cairo (StratoClim), A. Herber, J. Abbatt, R. Leaitch

(NETCARE). We also thank all aircraft crews, campaign teams and hangar staff (especially the staff of 120 ATW (Air Training Wing) at Kalamata), as well as the technical staff at MPIC for support during instrument development and operation during field campaigns.

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## Introduction

This document contains supplementary information, with the objective to give more background information on the data shown in the main article, to enable its interpretation to be scrutinised transparently. Firstly, the clustering methodology is explained in more detail, with the ~~It includes the~~ clustering parameters of the single particle data evaluation, and an associated ~~and the~~ uncertainty ~~estimation~~ estimated (S1). Secondly (S2), the mean mass spectra and vertical profiles of the meteoric-particle abundance fractions for each of the five UTLS campaigns are shown in Figures S1 to S5~~Individual clusters of particles are displayed (S2)~~. The data displayed in Figure 4 of the main text are shown for each of the individual missions in S3 (Figure S6). Section S4 (Figure S7) shows the $O_3$-$H_2O$ tracer-tracer plot for StratoClim 2016 which was not used in the main text. ~~Section S5 explains in more detail the correction for "missed" particles by using the hit rate of the mass spectrometer.~~ The calculation of the sedimentation velocity is explained in ~~S6~~S5. An example of sampling line loss calculation is presented in ~~S7~~S6. Section ~~S8~~ S7 shows the results of Mie calculations used to convert the calibration data to real stratospheric refractive indices. ~~Finally, some m~~More information on the detection of meteoric material in tropospheric particles at the Jungfraujoch is presented in ~~S9~~S8. Finally, Section ~~S10~~ S9 explains ~~the~~ how changing the threshold defining at what times stratospheric air was sampled affects the stratospheric proportions presented~~was derived~~.

## S1 Clustering Parameters

### Clustering algorithm and parameters

The individual bipolar mass spectra were sorted using the fuzzy c-means algorithm (Bezdek et al., 1984; Hinz et al., 1999), using the software CRISP that was written at MPIC (Klimach et al., 2010; Klimach, 2012; Roth et al., 2016). The parameters are given in Table S1. Cations were used because the meteoric material is best recognized in the cation spectrum ($Fe^+$, $Mg^+$). Preprocessing is done by taking each ion signal to the power of 0.5 to reduce the influence of the signal intensity. The mass spectra were normalized to their sum to reduce the influence of the total ion count per spectrum. Linear correlation was used for ~~chosen as~~the distance metric (defining "similarity" of the spectra), with ~~:~~ a perfect Pearson correlation ~~coefficient~~ (r = 1) meanings that two spectra are identical. The number of clusters was prescribed with 20. A set of starting cluster centers was chosen from the data set with the condition that these clusters have Pearson correlation coefficient smaller than 0.9. The fuzzifier (originally introduced as “weighting exponent” by Bezdek (1981) represents the fuzziness (blurring, defocusing) of the classification. "Fuzzy abort" parameter defines the convergence~~stopping~~ criterion of the algorithm, i.e. when the differences between subsequent cluster runs change by less than the chosen value, the algorithm ends.

### Variation of clustering parameters

To estimate the influence of the clustering parameters and the chosen sorting algorithm on the number of particles containing meteoric material, six additional different clustering runs were conducted for each UTLS

mission. The varied parameters are: number of clusters (10, 20, 30), initialization cluster difference (0.9, 0.7), fuzzyfier (1.3, 1.5), preprocessing (power = 0.5, none), and algorithm type (fuzzy c-means, k-means). Table S2 lists the different runs, the varied parameters and the resulting number of particles identified to contain ~~ing~~ meteori~~cological~~ material.

Criteria for selecting a certain cluster as "containing meteoric material" were 1) high cation signals of $Fe^+$ and $Mg^+$ (additionally allowing $Na^+$, $K^+$, $Al^+$), 2) anion signal at $HSO_4^-$ or cation signals at $S^+$, $SO_4^+$, $H_3SO_4^+$, 3) vertical profile showing increasing fractional abundance with increasing altitude, potential temperature, or potential vorticity.

In general, the standard deviation is below 6% of the mean value, and the chosen final clustering result using the parameters given in Table S1 (printed in bold) is very close to the mean value.

**S2 Individual cluster properties.**

Figures S1 through S5 show the particle clusters identified as "containing meteoric material" for each of the five UTLS aircraft missions. The cluster were selected following the criteria given in section S1.~~.~~

**S3 Theta-latitude histograms for individual aircraft missions.**

Figure S6 shows the theta-latitude histograms for each of ~~the~~ the five UTLS aircraft mission~~s, these~~. ~~All~~ data ~~were~~ merged to produce Figure 4 of the main text.

**S4 Tracer-tracer correlation for StratoClim 2016.**

The $O_3$ and $H_2O$ measurements during StratoClim 2016 did not cover the whole flight time of the three measurement flights. Thus, the data were not used in Figure 8 of the main text, but for completeness are shown here in Figure S7.

~~**S5 ALABAMA hit rate in stratosphere and troposphere.**~~

~~In section 3.5 of the main text we calculate the absolute number concentrations of particle containing meteoric material. For this, we use the hit rate of the mass spectrometer to estimate the number of "missed" particles. Figure S8 shows the relationship between hit rate and $O_3$ which allows for estimating the contribution of "invisible" pure $H_2SO_4$ particles in the stratosphere.~~

~~S6~~ S5 **Particle sedimentation in the lower stratosphere.**

The time scale for particle sedimentation was calculated as follows:

Pressure, temperature and viscosity of air were taken from the US Standard Atmosphere, using 100 m vertical resolution.

Mean free path ($\lambda$), Knudsen number ($Kn$), Cunningham correction ($C_C$) and terminal settling velocity ($V_{TS}$) were calculated using the following equations ((Hinds, 1999; Seinfeld and Pandis, 2006)):

$$\lambda = \frac{1}{\sqrt{2}\pi d_m^2 N}, \quad \text{(S1)}$$

with $d_m$ = collision diameter of air molecules ($3.7\times10^{-10}$ m), and $N$ = number density of air molecules,

$$Kn = \frac{2\lambda}{d} \quad \text{(S2)}$$

with $d$ = particle diameter,

$$C_C = 1 + Kn\left(\alpha + \beta e^{-\frac{\gamma}{Kn}}\right), \quad \text{(S3)}$$

with $\alpha$ = 1.155, $\beta$ = 0.471, $\gamma$ = 0.596 (Allan and Raabe, 1982)

$$V_{TS} = \frac{\varrho\, d^2\, g\, C_C}{18\,\eta}, \quad \text{(S4)}$$

with $\varrho$ = particle density, $g$ = acceleration of gravity, $\eta$ = viscosity of air.

The terminal settling velocity was calculated for pure $H_2SO_4$ particles ($\varrho$ = 1.83 g cm$^{-3}$) and pure olivine particles ($\varrho$ = 3.30 g cm$^{-3}$), assuming spherical particle shape. Figure ~~S9~~ S8 shows the terminal settling velocity as a function of altitude.

**S~~6~~7 Sampling line transmission efficiency.**

The sampling line transmission efficiency was calculated here as an example for the configuration of ML-CIRRUS (ALABAMA operated on the HALO aircraft). The ¼" stainless steel sampling line that connected the HALO aerosol submicrometer inlet (HASI, Wendisch et al. (2016); Andreae et al. (2018)) line had a total length of 2.9 m with several bends, horizontal and vertical sections. The calculations were done with a modified version of the Particle Loss Calculator (PLC) that was originally described in von der Weiden et al. (2009). The modified version allows for including the sampling line pressure. The results are shown in Figure ~~S10~~S9.

**S~~7~~8 Mie calculations for stratospheric aerosol for the optical particle spectrometers OPC 1.129 and UHSAS.**

The response of the optical particle spectrometers OPC 1.129 and UHSAS for stratospheric aerosol particles was calculated using an in-house written software (Vetter, 2004) following the algorithms described in Bohren and Huffmann (1983). The OPC 1.129 uses a laser wavelength of 655 nm. The scattered light is collected under 90° with an angular range of 60° (i.e. 60° – 120°). The UHSAS uses a laser wavelength of 1054 nm and collects the scattered light in an angular range between 22° and 158°. The refractive index for PSL, $m$ = 1.59, was taken from Heim et al. (2008). The refractive index range ($m$ = 1.43 – 1.45) for lower stratospheric aerosol was taken from Yue et al. (1994).

The results (Figure S1~~1~~0) show that the lower size cut-off of the OPC shifts from 250 nm to about 285 nm when the refractive index for stratospheric aerosol is used. For the UHSAS, the ~~size channel with~~ lower cut-off of 180 nm (calibrated by PSL) increases ~~corresponds~~ to 200 nm ~~for stratospheric aerosol particles~~.

**S~~8~~9 Jungfraujoch backtrajectories, ozone mixing ratio, and meteoric particle fraction.**

We inspected backtrajectories to obtain more information on the origin of the meteoric particles detected during the INUIT-2017 campaign at the Jungfraujoch station (3600 m altitude). For this, HYSPLIT (Stein et al., 2015) back trajectories were calculated for 120 – 143 hours using the GDAS 0.5 degree data set (NCEP meteorological re-analysis, Saha et al., 2010). We chose 3600 m a.s.l. as a starting point, with~~.~~ 27 trajectories were started per hour, using the trajectory ensemble option. All trajectory data points were stratified into~~binned in~~ altitude and latitude bins and the number of trajectory points per grid is plotted in Figure ~~S12~~ S11 a) and b). Panel ~~-~~c) of Figure S1~~1~~2 shows the time series of the fraction of meteoric particles detected along with the $O_3$ mixing ratio. The fraction of meteoric particles is found to be highest between the 18th and 21st of February, a period during which the air mass spent more time at higher altitudes and latitudes before arriving at the Jungfraujoch. A similar but less pronounced feature is found between 15th and 17th of February. These findings support the conclusion that the origin of this particle type is at higher altitudes. The dependence on latitude can be explained by the fact that mixing between stratosphere and troposphere is stronger at mid-latitudes (see Figure 8 in the main part) than in the tropics. Thus, we would expect to see a higher meteoric particle fraction when the air masses have experienced higher latitudes and altitudes, which is confirmed by Figure S1~~1~~2. Furthermore, a stratospheric origin is supported by the ozone trend in Panel c). The time trend of the meteoric particles follows closely the ozone time series, and even small-scale features (e.g. Feb. 09, Feb. 11, Feb. 16, and Feb. 22) are clearly visible in both time series.

**S~~9~~10 Number of particle mass spectra in the stratosphere**

Table S3 shows the number of particle mass spectra measured~~recorded~~ in the stratosphere, for all particles and for meteoric particles. To test the sensitivity of the calculations, w~~W~~e used different definitions of the tropopause (PV > 2 PVU, PV > 4 PVU, $O_3$ > 150 ppbv, and pot. temp. > 380 K. The la~~e~~tter criterion could only be applied to the StratoClim data sets, because only the Geophysica reaches to 380 K. The ozone criterion could not be applied to the StratoClim 2016 data set due to low data coverage. As a final result, we decided to use PV > 4 PVU as ~~a~~ the threshold for Table 1 in the main text.

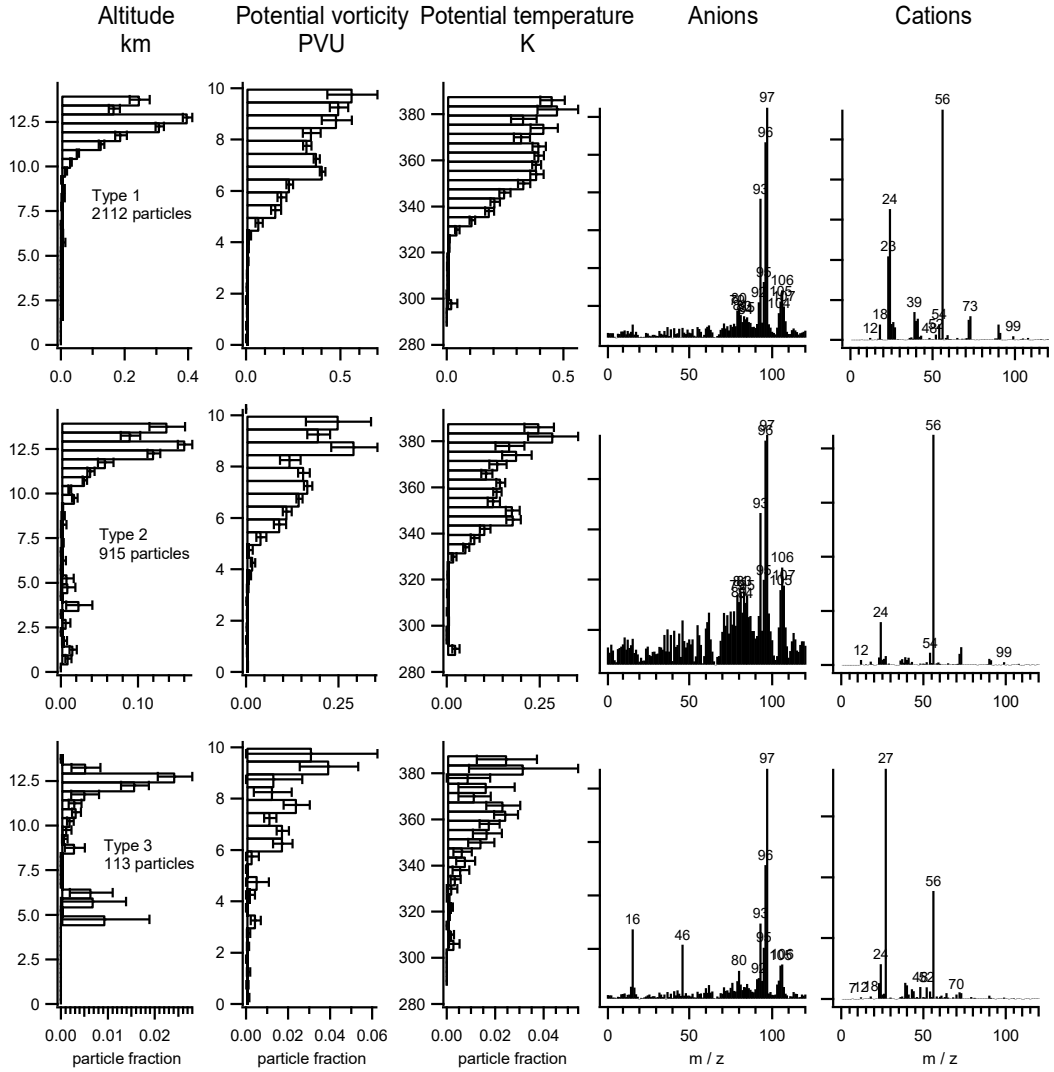



**Figure S1. Vertical profiles and mean mass spectra of all clusters from the ML-CIRRUS 2014 data set interpreted as particles containing meteoric material. Note that the negative mass spectra (anions) are noisy, because all anion spectra of each cluster type were averaged for this display, also those where no anion signal was obtained.**

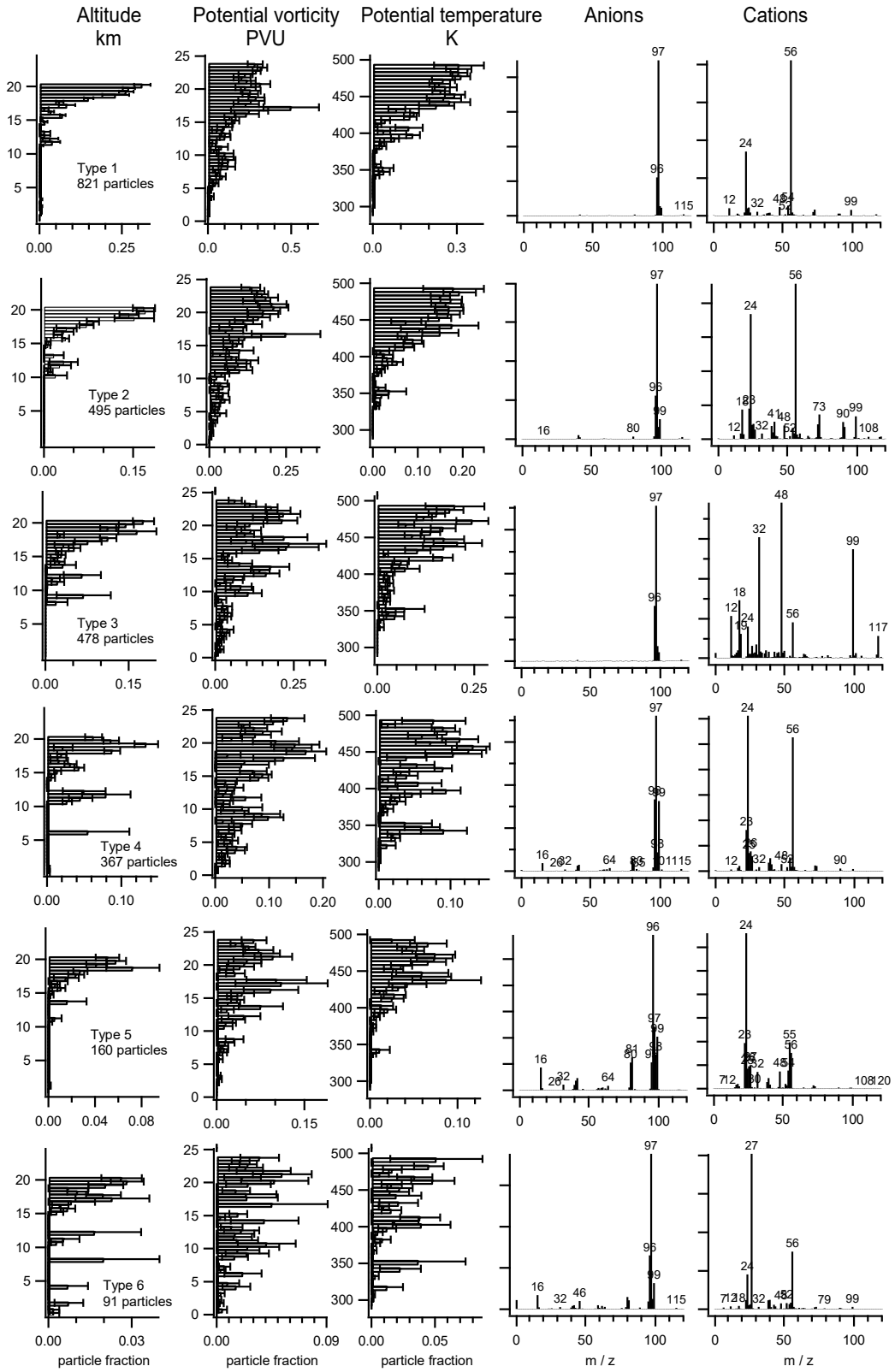



**Figure S2. Vertical profiles and mean mass spectra of all clusters from the StratoClim 2016 data set interpreted as particles containing meteoric material.**

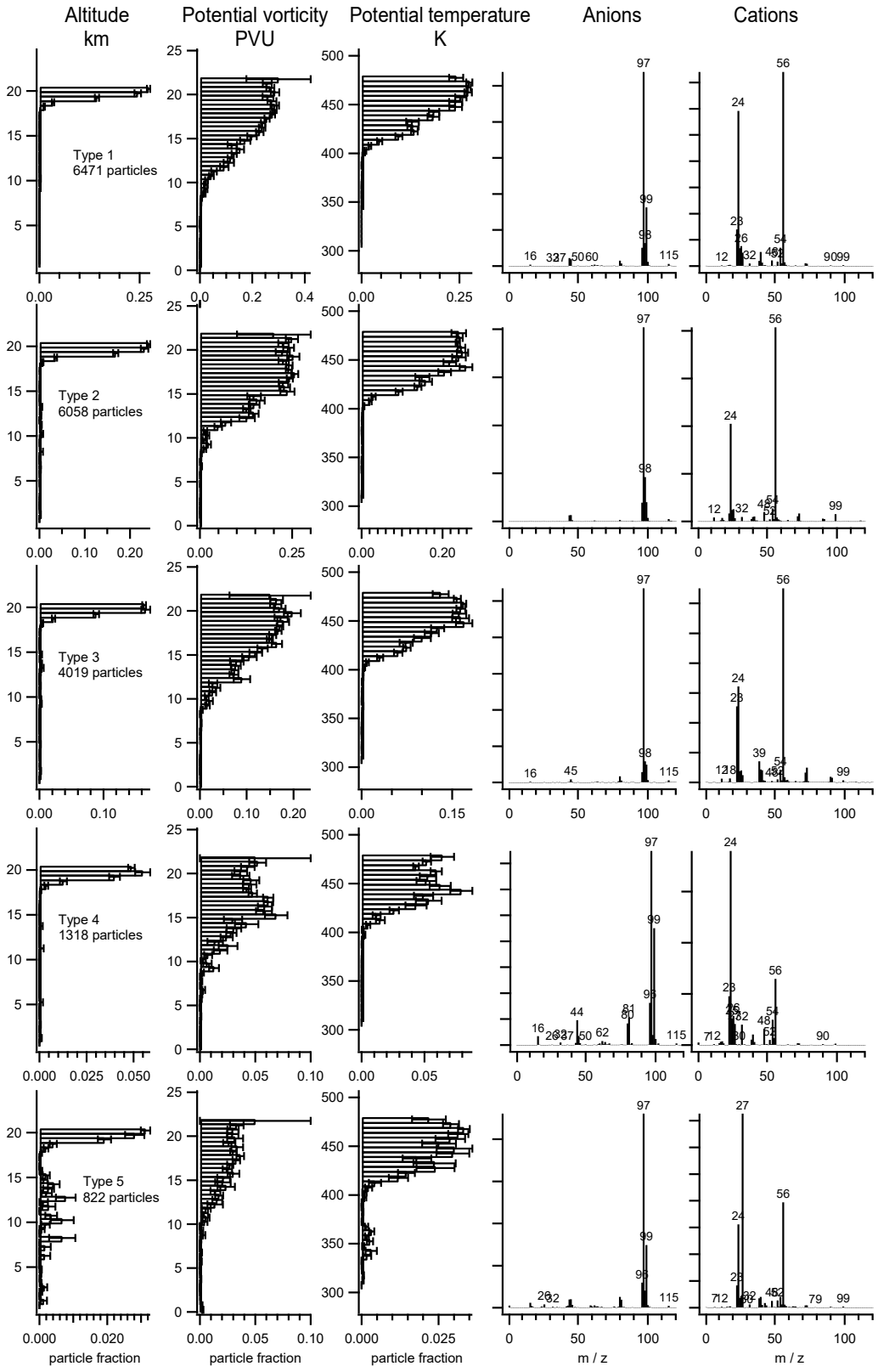



**Figure S3. Vertical profiles and mean mass spectra of all clusters from the StratoClim 2017 data set interpreted as particles containing meteoric material.**

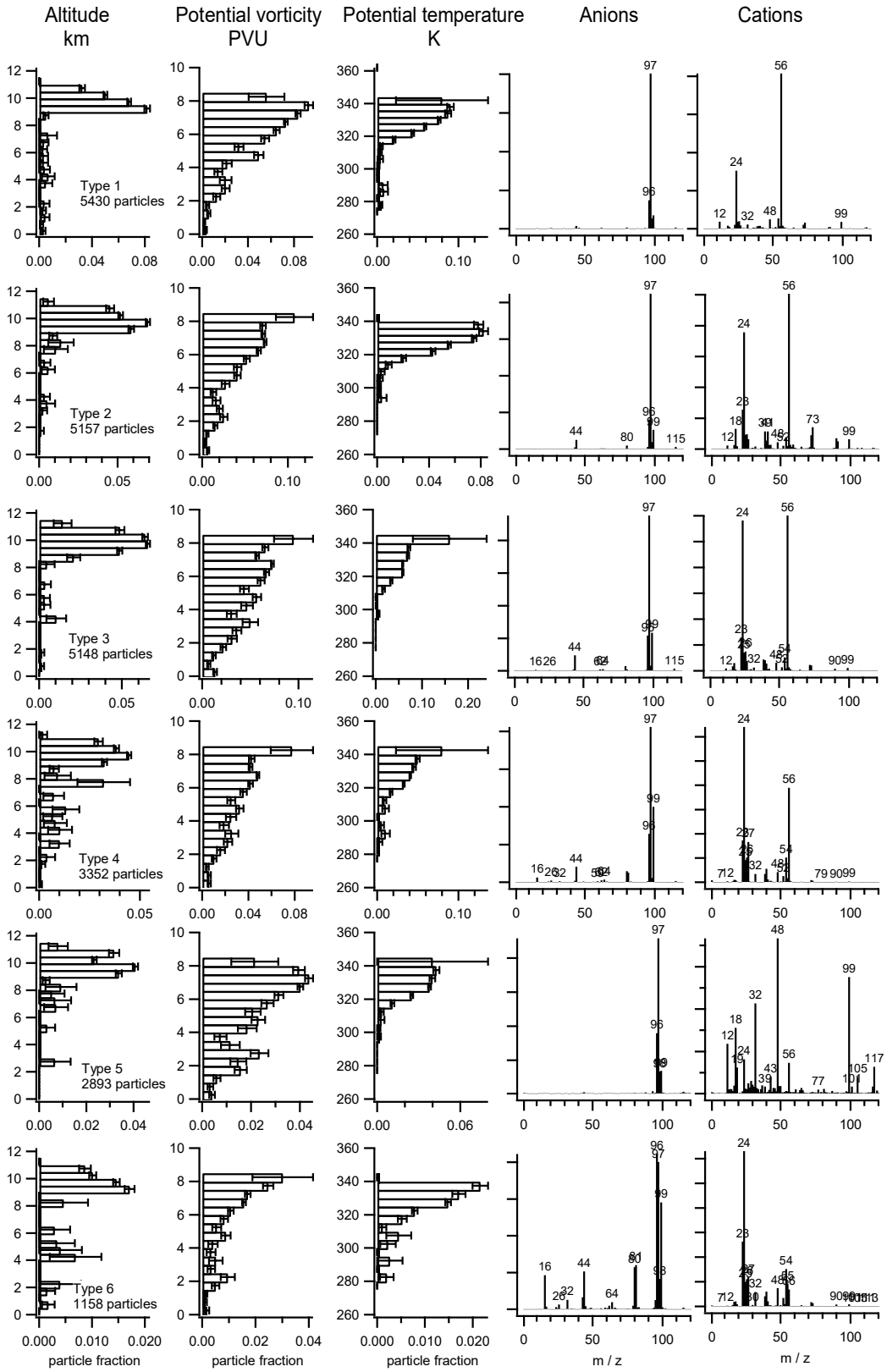



**Figure S4. Vertical profiles and mean mass spectra of all clusters from the ND-MAX/ECLIF-2 data set interpreted as particles containing meteoric material.**

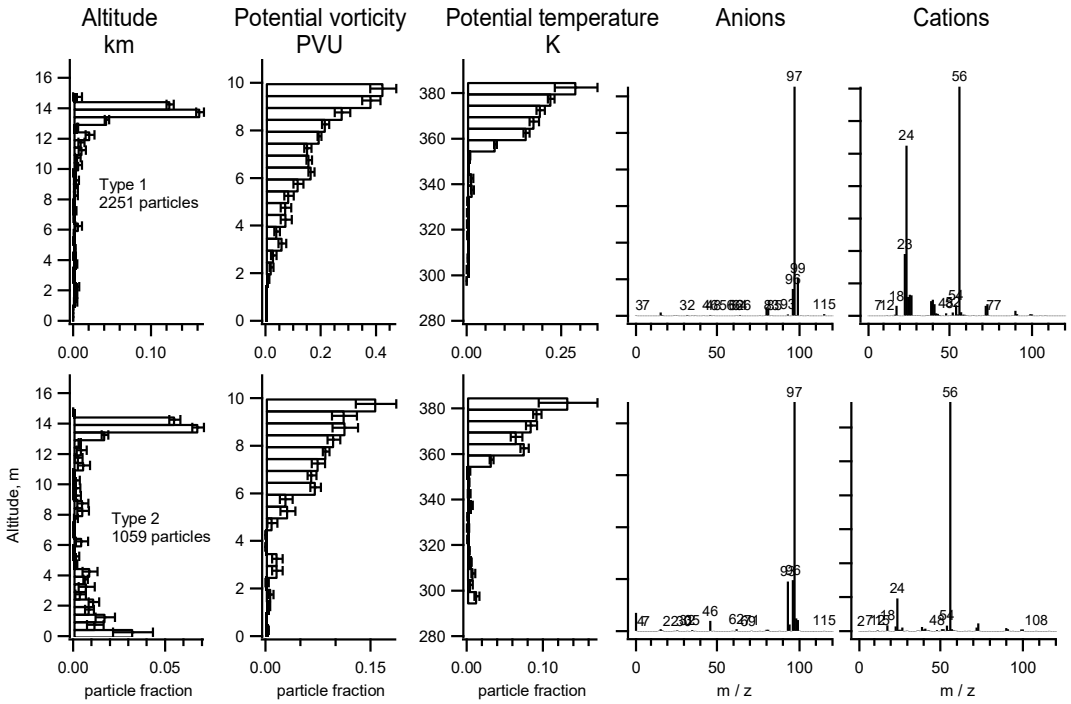



**Figure S5. Vertical profiles and mean mass spectra of all clusters from the CAFE-Africa data set interpreted as particles containing meteoric material.**

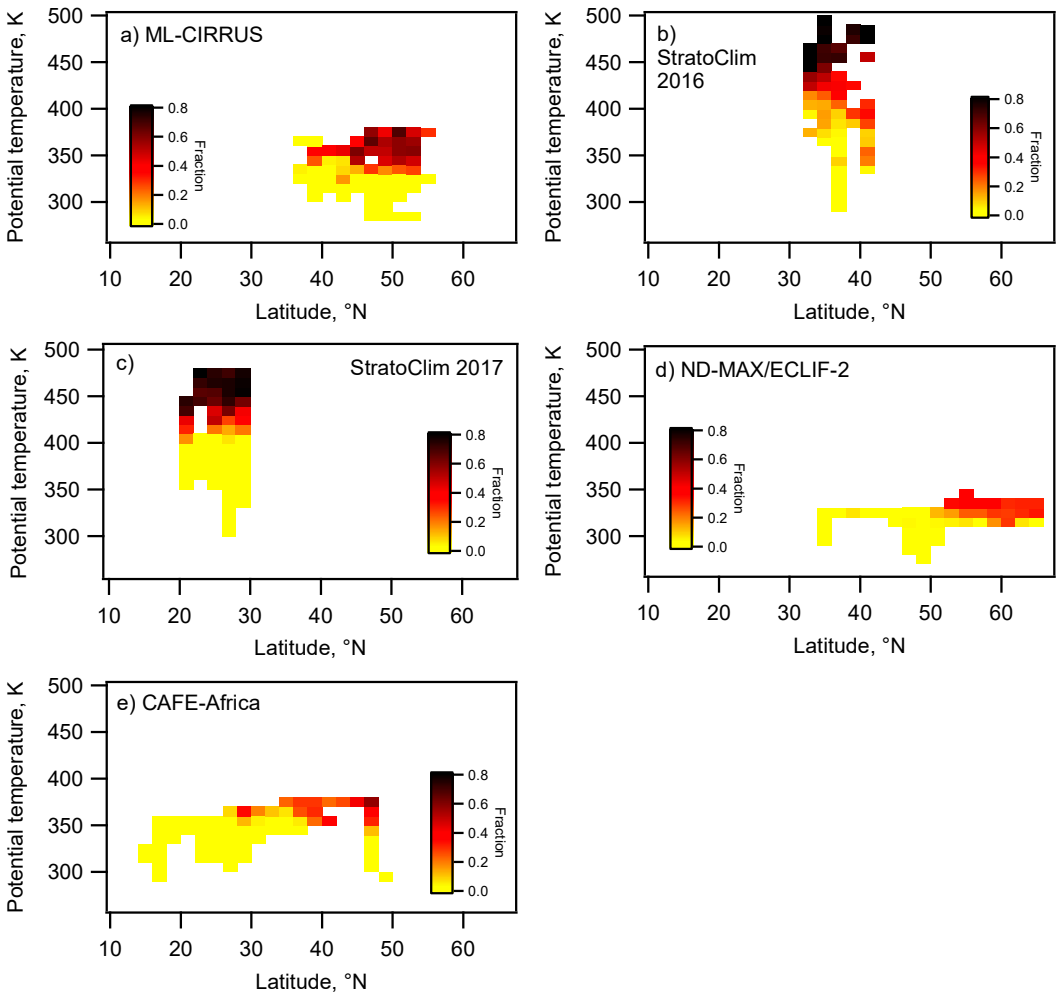



**Figure S6. Fraction of meteoric particles as function of potential temperature and latitude for the individual missions: a) ML-CIRRUS, b) StratoClim 2016, c) StratoClim 2017, d) ND-MAX-ECLIF-2, e) CAFE-Africa.**

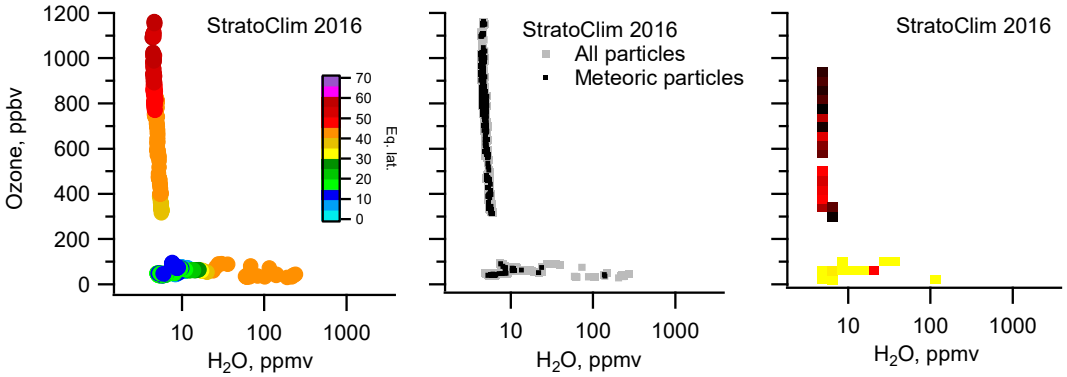
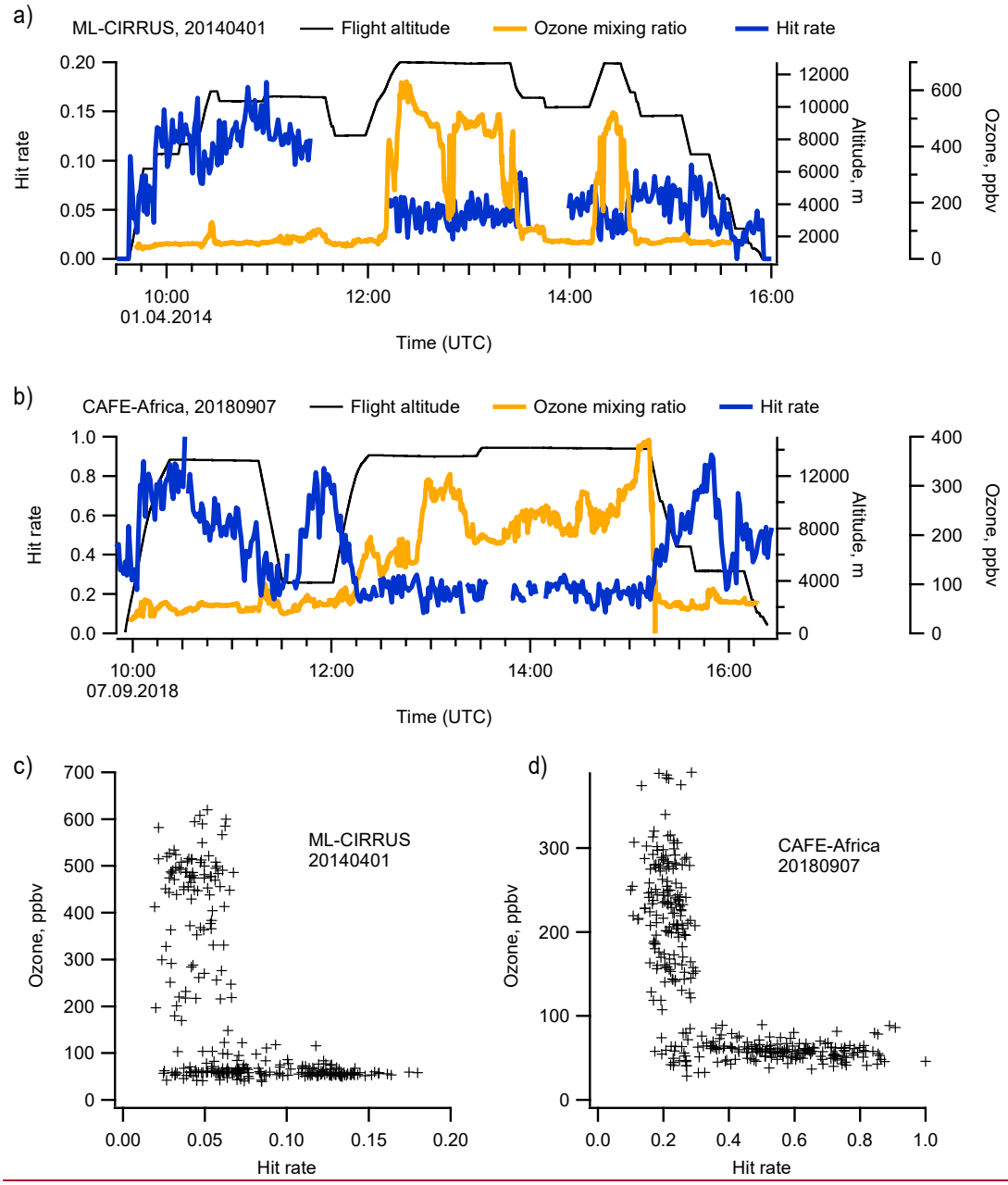



**Figure S7. Tracer-tracer correlation for StratoClim 2016. As the instruments were not fully operative during the whole flight time of the three flights of StratoClim 2016, several gaps in the data prevent the analysis of cross-tropopause transport for this mission.**




~~**Figure S8. Hit rate (mass spectra per laser shot during time intervals of one minute) of ALABAMA during a) a flight on April 01, 2014 (ML_CIRRUS) and b) a flight on Sept 07, 2018 (CAFE_Africa) as a function of time along with ozone mixing ratio and flight altitude. c) + d): Hit rate versus ozone mixing ratio. Dividing between troposphere and stratosphere at 150 ppbv ozone (ML_CIRRUS) and 100 ppb ozone (CAFE_Africa), respectively, yields an average hit rate in the stratosphere of 0.044 ± 0.012 (ML_CIRRUS) and 0.21 ± 0.04 (CAFE_Africa). For the troposphere, we obtain 0.091 ± 0.037 (ML_CIRRUS) and 0.54 ± 0.18 (CAFE_Africa).**~~

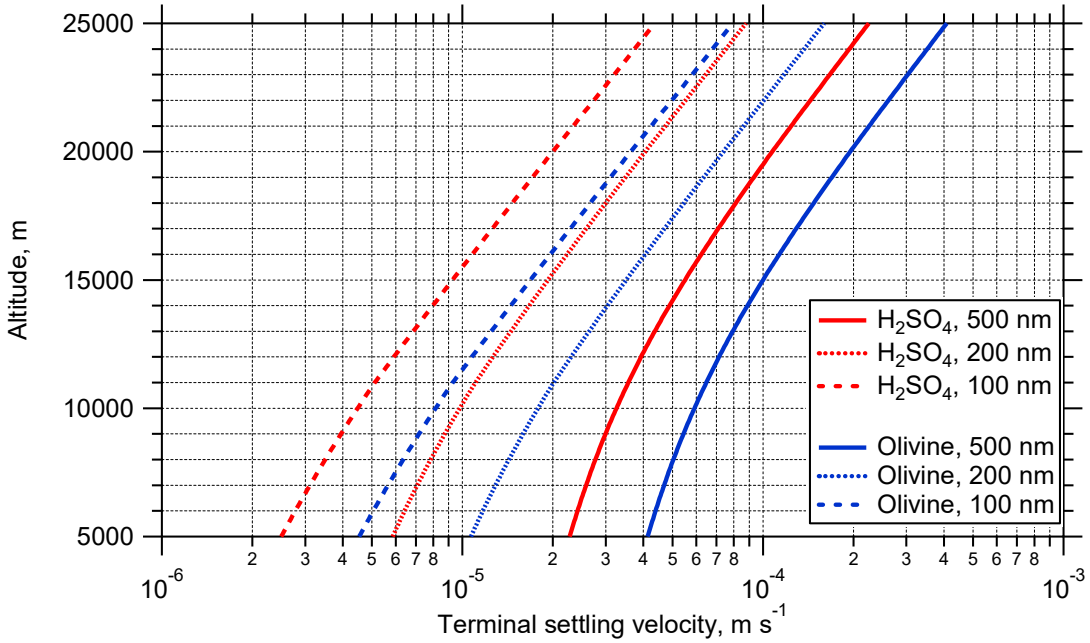



**Figure S89. Terminal settling velocity for $H_2SO_4$ and olivine particles of 100, 200, and 500 nm volume equivalent diameter ($d_{ve}$).**

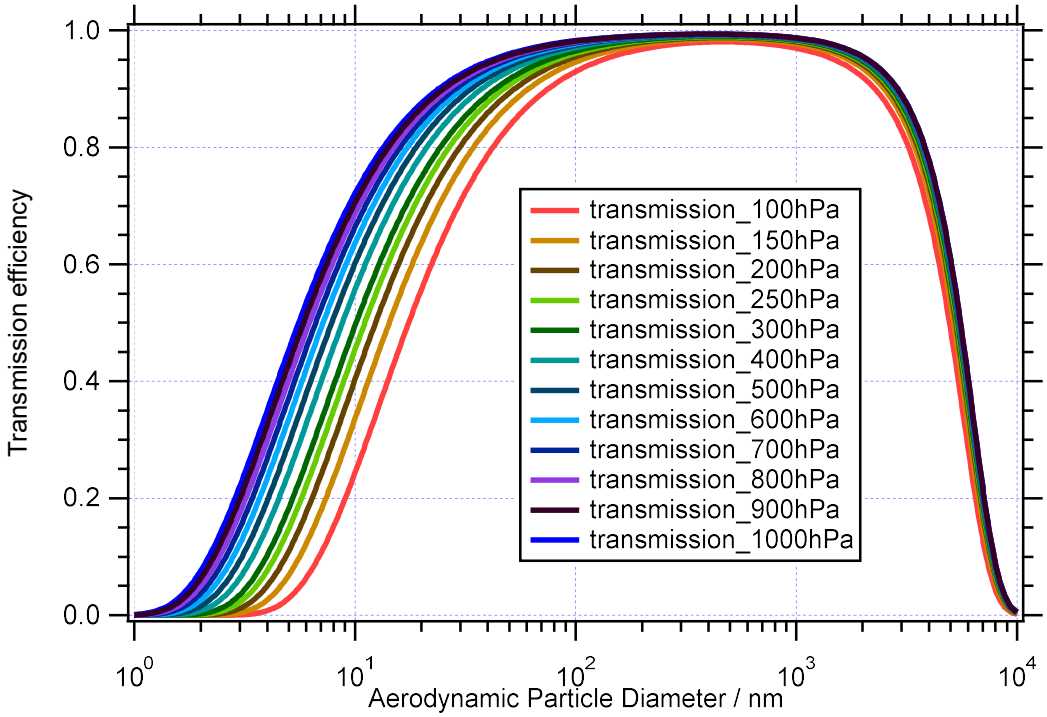



**Figure S9~~10~~. Transmission curves for the sampling line used in ML-CIRRUS that connected the HALO aerosol submicrometer inlet (HASI) to the OPC 1.129 in the ALABAMA rack.**

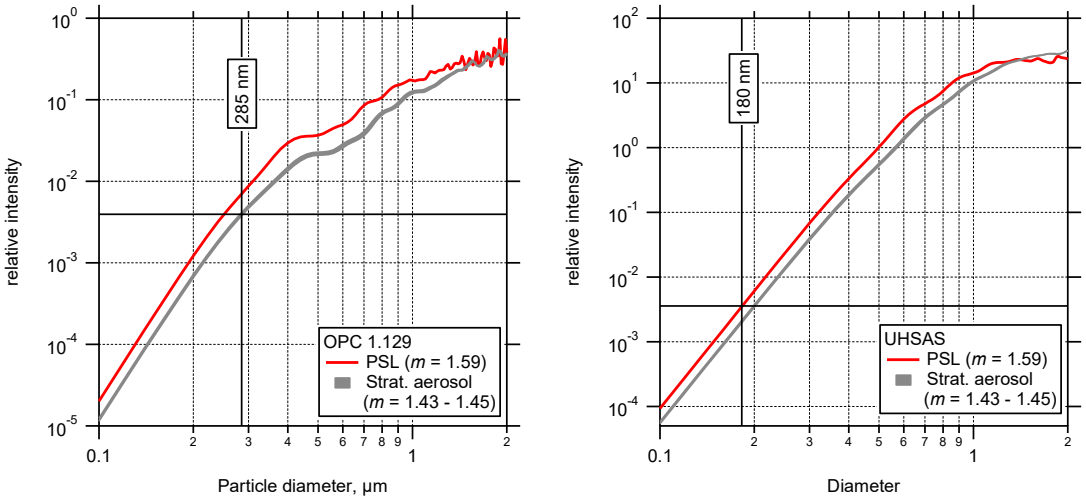



**Figure S101. Relative intensity at the detector calculated for the OPC (Grimm 1.129, "Sky-OPC") and for the UHSAS, for PSL particles and for stratospheric particles.**

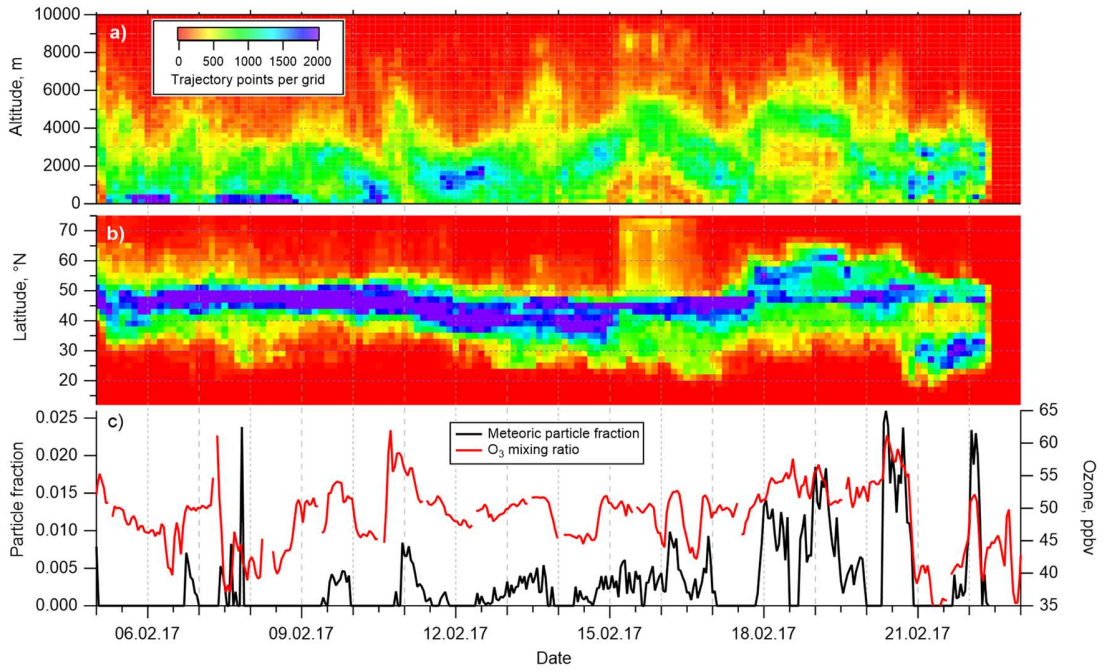



**Figure S121. Backtrajectory information for the INUIT-JFJ 2017 field campaign at Jungfraujoch. For each hour, 27 backtrajectories were started using the trajectory ensemble option and were followed for 120 – 143 h using HYSPLIT [Stein et al., 2015] with the GADS 0.5 degree data set. Panel a) shows the number of points during which the trajectories resided in the respective altitude and time grid, Panel b) the same but for latitude. Panel c) shows the number fraction of the meteoric particles along with ozone mixing ratio.**

| | |
|---|---|
| Ion type | Cations |
| Preprocessing | Power each m/z by 0.5 |
| Normalization | Sum |
| Distance metric | Correlation |
| Initialization | Different startclusters: |
| Number of clusters | 20 |
| Cluster difference | 0.9 |
| Fuzzifier | 1.3 |
| Fuzzy abort | 1e-5 |

Table S1. Clustering parameters used for the final analysis.

| Project | Cluster algorithm | Number of clusters | Cluster diff | Fuzzifier | Pre-processing | Number of "meteoric" particles |
|---|---|---|---|---|---|---|
| ML-CIRRUS | Fuzzy c-means | 10 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 3080 |
| | **Fuzzy c-means** | **20** | **0.9** | **1.3** | **$(m/z)^{0.5}$** | **3140** |
| | Fuzzy c-means | 30 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 3136 |
| | Fuzzy c-means | 20 | 0.7 | 1.3 | $(m/z)^{0.5}$ | 2931 |
| | Fuzzy c-mean | 20 | 0.9 | 1.5 | $(m/z)^{0.5}$ | 3136 |
| | Fuzzy c-means | 20 | 0.9 | 1.3 | none | 3051 |
| | k-means | 20 | 0.9 | N/A | $(m/z)^{0.5}$ | 3247 |
| Mean ± StdDev | | | | | | 3103 ± 90 |
| | | | | | | |
| StratoClim 2016 | Fuzzy c-means | 10 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 2357 |
| | **Fuzzy c-means** | **20** | **0.9** | **1.3** | **$(m/z)^{0.5}$** | **2412** |
| | Fuzzy c-means | 30 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 2679 |
| | Fuzzy c-means | 20 | 0.7 | 1.3 | $(m/z)^{0.5}$ | 2376 |
| | Fuzzy c-mean | 20 | 0.9 | 1.5 | $(m/z)^{0.5}$ | 2567 |
| | Fuzzy c-means | 20 | 0.9 | 1.3 | none | 2618 |
| | k-means | 20 | 0.9 | N/A | $(m/z)^{0.5}$ | 2570 |
| Mean ± StdDev | | | | | | 2511 ± 118 |
| | | | | | | |
| StratoClim 2017 | Fuzzy c-means | 10 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 18355 |
| | **Fuzzy c-means** | **20** | **0.9** | **1.3** | **$(m/z)^{0.5}$** | **18688** |
| | Fuzzy c-means | 30 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 19700 |
| | Fuzzy c-means | 20 | 0.7 | 1.3 | $(m/z)^{0.5}$ | 18688 |
| | Fuzzy c-mean | 20 | 0.9 | 1.5 | $(m/z)^{0.5}$ | 18459 |
| | Fuzzy c-means | 20 | 0.9 | 1.3 | none | 21235 |
| | k-means | 20 | 0.9 | N/A | $(m/z)^{0.5}$ | 20215 |
| Mean ± StdDev | | | | | | 19334 ± 1006 |
| | | | | | | |
| ND-MAX/ ECLIF-2 | Fuzzy c-means | 10 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 20141 |
| | **Fuzzy c-means** | **20** | **0.9** | **1.3** | **$(m/z)^{0.5}$** | **23138** |
| | Fuzzy c-means | 30 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 21883 |
| | Fuzzy c-means | 20 | 0.7 | 1.3 | $(m/z)^{0.5}$ | 21681 |
| | Fuzzy c-mean | 20 | 0.9 | 1.5 | $(m/z)^{0.5}$ | 21126 |
| | Fuzzy c-means | 20 | 0.9 | 1.3 | none | 21752 |
| | k-means | 20 | 0.9 | N/A | $(m/z)^{0.5}$ | 18998 |
| Mean ± StdDev | | | | | | 21245 ± 1237 |
| | | | | | | |
| CAFE-Africa | Fuzzy c-means | 10 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 3325 |
| | **Fuzzy c-means** | **20** | **0.9** | **1.3** | **$(m/z)^{0.5}$** | **3310** |
| | Fuzzy c-means | 30 | 0.9 | 1.3 | $(m/z)^{0.5}$ | 3290 |
| | Fuzzy c-means | 20 | 0.7 | 1.3 | $(m/z)^{0.5}$ | 3194 |
| | Fuzzy c-mean | 20 | 0.9 | 1.5 | $(m/z)^{0.5}$ | 3281 |
| | Fuzzy c-means | 20 | 0.9 | 1.3 | none | 3287 |
| | k-means | 20 | 0.9 | N/A | $(m/z)^{0.5}$ | 3515 |
| Mean ± StdDev | | | | | | 3314 ± 90 |
| | | | | | | |

Table S2. Variations of clustering parameters. The inferred number of particles containing meteoric material is given in the last column. Other parameters were kept as in Table S1.

a) All particles

| | PV > 2 | PV > 4 | O3 > 150 | Theta > 380 |
|---|---|---|---|---|
| ML-CIRRUS | 13029 | 6509 | 6174 | N/A |
| SC16 | 6662 | 5092 | N/A | 4874 |
| SC17 | 76856 | 51599 | 41146 | 57109 |
| NDMAX | 78454 | 73367 | 72923 | N/A |
| CAFE | 12161 | 10771 | 9441 | N/A |

b) Meteoric particles

| | PV > 2 | PV > 4 | O3 > 150 | Theta > 380 |
|---|---|---|---|---|
| ML-CIRRUS | 3063 | 2986 | 2477 | N/A |
| SC16 | 2363 | 2271 | N/A | 2238 |
| SC17 | 18487 | 18421 | 18016 | 18450 |
| NDMAX | 22626 | 22050 | 22104 | N/A |
| CAFE | 2946 | 2882 | 2789 | N/A |

c) Proportion

| | PV > 2 | PV > 4 | O3 > 150 | Theta > 380 |
|---|---|---|---|---|
| ML-CIRRUS | 0.235 | 0.459 | 0.401 | N/A |
| SC16 | 0.355 | 0.446 | N/A | 0.459 |
| SC17 | 0.241 | 0.357 | 0.438 | 0.323 |
| NDMAX | 0.228 | 0.301 | 0.303 | N/A |
| CAFE | 0.242 | 0.268 | 0.295 | N/A |

Table S3. a) Number of analyzed particles in the stratosphere; b) number of meteoric particles in the stratosphere, c) proportion (numbers in b) divided by numbers in a)). Different criteria were used to define the tropopause. For Table 1 and Figure 6 in the main text, PV > 4 was selected for the following reasons: to avoid the mixing regime at mid-latitudes and to match the tropical 380 K tropopause definition (see also (Ploeger et al., 2015)).

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