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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