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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Aircraft-based observation of meteoric material in lower stratospheric aerosol particles between 15 and 68° N" *by* Johannes Schneider et al.

Anonymous Referee #1

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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.

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.

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.

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". 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. 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.

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 CAFÉ-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.

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.

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.

minor and typographical changes:

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

Page 14 line 330 should read "boundary between troposphere and stratosphere"

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.

P21 line 511 "between" should read "above"

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"?

P25 Line 585 change to "particles containing"

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.

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

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

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