Comment on Schütze et al. (Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-278, 2017)

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This article has the potential to be a useful addition to the understanding of stratospheric aerosol, particularly since there are relatively few capture and return samples with statistics on this level. Whilst the conclusions they are able to draw are limited, publication of such data is vital in facilitating future understanding. I feel that the authors have missed or omitted a section of the literature which, once considered, can both add to the understanding of the results and increase the potential audience of the article. Below are a number of specific comments on language, formatting and content.

The authors provide a good introduction to the current and historical field. I was surprised to see no reference to the recent and thorough review of Kremser et al. (2016), who summarised some of the studies mentioned in the introduction of this work and other related topics.

Page 4 line 2; change “extend” to read “extent”.

The analysis of images for structure of carbonaceous material is interesting. Was electron diffraction data recorded for any samples?

Page 5 line 8; reformat 5×10^{-3}.

Page 5 line 27; amend to “too close to”

Page 5 line 30 onwards; sentence is hard to understand. Perhaps “Any particle which showed no signs of destruction or morphological change was taken to be non-volatile. Particles which changed under the electron beam were deemed volatile, allowing quantification of the fraction of aerosol which is volatile.”

Section 4.1; I believe this section would benefit from also discussing the size ranges of the various particles. For example Ebert et al. (2016) discuss mainly particles of radius greater than 500 nm, which have metallic or meteoritic composition. In that study the smaller size fraction is described as being largely carbonaceous material in sulfate liquid droplets, similar to the findings of this study.

Page 12 line 1; change to “the particles described above matches the refractory…”

Possibility that particles have an extraterrestrial origin; this section makes a good comparison between measurements of extraterrestrial material and the particles observed in the stratosphere. What is lacking is any discussion of the process which occur as a result of frictional heating during atmospheric entry. There is currently some discussion of whether unablated meteoric material will sediment too rapidly to be found in the stratosphere (Carrillo-Sánchez et al., 2016), or whether significant fragmentation of ablating meteorites could lead to smaller aerosol with longer atmospheric lifetimes (Subasinghe et al., 2016). Considering these processes in the light of the results presented here would broaden the appeal of the current results to a wider audience and add significantly to the conclusions the authors are able to draw from their data.

Regarding ablation: The fact that the three metals discussed; Ni, Fe and Cr; have ratios significantly different than their chondritic abundances has rather more interesting implications when considered with respect to the ablation process. Since in interplanetary dust Ni is largely contained in relatively volatile metal phases (melting points around 1200-1500 K), Fe is spread between volatile metallic and more refractory silicate phases (melting point >1800 K) and Cr is contained in the less volatile silicates (Bunch and Olsen, 1975), the three elements will ablate rather differently (Gómez-Martín et al., 2017). The relative volatility of Ni is therefore reconcilable with the larger Ni/Fe ratio measured here and does not rule out an extraterrestrial source, but the high Cr/Fe and Cr/Ni suggest that Cr at least has a terrestrial source, since if anything Cr should ablate less completely than the other elements.

Regarding fragmentation: This is hypothesised to happen by evaporation of volatile phases such as iron sulfides and amorphous carbonaceous material (ordered graphitic material would be much more refractory). It may be reasonable as a result that the metal bearing silicates would remain in larger particles which have very short lifetimes in the stratosphere, but carbonaceous material and some additional Fe is atmospherically available as a result.

The question could possibly be more constructively phrased in another way. Since we know that meteoric ablation occurs and meteoric smoke forms, why is it not unequivocally observed in these measurements? Indeed numerical modelling of MSPs suggests that they should be observable in this size range (Bardeen et al., 2008). Could it be that nucleation, growth and sedimentation of crystalline PSC has removed meteoric material? What implications would the partial dissolution of metals have on these measurements? Could dissolution, precipitation and agglomeration in liquid droplets cause more rapid growth of MSP compared to model predictions?

These comments also inherently include the issue of sample aging, which both anonymous reviewers rightly mention. Having some experience of electron microscopy, I suspect that this statistical detail could only be reached from measurements which would take several years to make. In addition to the reviewer's comments then, the possibility should be considered that some samples have aged more than others.

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