Comments by A. James

Authors: We gratefully acknowledge the suggestions and advices that uncovered remaining deficits of previously submitted article. We believe that the effort of all involved referees contributed crucially to the improvement of this paper.

A. James: As mentioned by one reviewer, it is important to consider modelling studies of meteoric smoke when interpreting the data.
The heated COPAS inlet has a lower particle size cutoff at 10 nm. However, Bardeen (2008, J. Geophys. Res.) predicted that a significant amount of the meteoric mass will be in smaller (_3 nm) particles at these altitudes. This is mentioned in sections 4.1 and 5.1, however the impact of dissolution of such particles is not discussed. MSP will dissolve when entrained in acidic liquid aerosol (Saunders, 2012, Atmos. Chem. Phys.). Measurements of midlatitude stratospheric aerosol have shown that Junge layer sulfuric acid droplets contain 0.5 wt% iron (Cziczo, 2001, Science). I estimate that 0.5 wt% of iron condensing from a solution droplet of 500 nm radius to an iron sulfate crystal could not have an equivalent spherical radius of more than 2 nm. However, the 'half of particles contain 1 wt% iron' number is from midlatitudes and could be higher (due to mesospheric influx) or lower (due to growth of Junge layer aerosol at colder temperatures). The 6 nm unheated channel might be expected to pick up some of this material, but not if MSP were rapidly coated with acids at higher altitudes. This leads to the possibility that a significant portion of meteoric smoke will not be detected by the COPAS instrument, and should be discussed in the paper.

> **Author's reply to Comment:** We agree with A. James in that point that model studies like Bardeen et al. (2008) and laboratory investigations as described by Saunders et al. (2012) are currently not considered in the manuscript and in correspondence to the reply to Referee 2 the revised paper version will be completed in this sense. We furthermore do not disagree with the referenced studies by Cziczo et al. (2001) and the conclusive estimate by A. James out of this. However, as stated, their samples from the mid-latitude lowermost stratosphere, where the measurements occurred, may not represent the refractory content of the stratospheric aerosol, in general.
> However, accounting also for the studies of Bardeen et al. (2008) in the Arctic stratosphere the MSP may be dominated by sub-10nm particles (down to 30 km altitude) but larger MSP may also exist. If individual MSP are activated when entering H_2SO_4 saturated regime from aloft then the largest MSP are likely activated first, and the smallest (if not immediately activated) are most likely incorporated by H_2SO_4 particles very quickly. Due to different sedimentation speed as a function of size, for a polydisperse aerosol population the coagulation probability is increased. Thus, the ultrafine particles will be incorporated into larger particles and the peak of the size distribution may move towards larger particle sizes.
> Dissolution of MSPs, or parts of them, may occur in the liquid acidic phase as well. What happens if a H_2SO_4 particle evaporates that includes one or more initially individual MSP? The surface tension of H_2SO_4 is quite high which, if the H_2SO_4 hull contracts, likely causes the individual substances (dissolved ions, MSP, other refractory material) to adhere together. Most likely the refractory remnant will be one residual, either from one individual precursor or as a composition of two or more individuals. In any case we try to clarify all of this in the revised paper also in correspondence to the replies to the other referees.

A. James: The density assumption for meteoric material seems reasonable, however there is also significant terrestrial material in the stratosphere. Additionally, if the refractory aerosol detected is coming out of solution when a liquid aerosol evaporates, the salt density would be higher. (iron sulphate hydrates have densities from 1.8-3 g cm^{-3}).

> **Author's reply to Comment:** As in the revised version the study of Bardeen et al.(2008) will be considered the recalculation of our estimate (with predominantly smaller

particles) will also account for the possibly larger material densities. The recalculation includes a range of mean material densities between 1 and 3 g cm^{-3}, and 2 g cm^{-3} is used as the mean of material densities.

A. James: MIF (section 5.4):
The discussion of MSP dynamics given in one of the independent reviews, combined with the possibilities of higher density residuals and a larger contribution from smaller MSP, could lead to an even greater discrepancy with the MIF.On the other hand, aerosol (such as Junge layer droplets containing eg. Terrestrial dust or soot) may penetrate the vortex more easily than gasses (such as N2O). ie. There may be a significant contribution of 'non-mesospheric' non-volatile aerosol being counted as meteoric influx. This could have a large impact on the estimate produced for the MIF. A further study is mentioned which will describe the chemical nature of the refractory material. Hopefully this will shed some light on the origin of the aerosol. Without accounting for other sources of refractory aerosol this study can only be considered as an upper limit to the MIF.

Author's reply to Comment: We clearly stated that our closure between detected aerosol number concentration and total aerosol mass within the vortex of the Arctic is a "highly uncertain" estimate. We agree that some emphasis should be put on how our results can be interpreted with respect to different particle's origin and transport processes within the atmosphere. This is envisaged in the revised paper version as far as possible based on the CPC measurements.