Anonymous Referee #3:

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.

Referee #3: Major comments;
Overall: The writing throughout the paper is very hard to follow and overly complicated. Much of this appears to be related to the authors writing style which should be simplified to promote readability. To be honest, I found myself reading and rereading sections trying to be sure that my interpretation of the authors' intent was correct. I shouldn't have to do that.

> **Author's reply to Comment:** Following the recommendation of other referees, this article is comprehensively restructured and rephrased. We also involved a native English speaker for the proof-reading and improving readability.

Referee #3: Overall: There is a tendency to write portions of the text as if the sulfate/volatile aerosol is a separate component of the aerosol from the non-volatile refractory portion. I suspect that this is just some inexact wording but it is a bit disconcerting nonetheless.

> **Author's reply to Comment:** Indeed, once the refractory cores are detected after having passed the heated aerosol line the refractory compounds are separated from the sulfate/volatile aerosol. In fact the "component separation" seems to be more a consequence of the measurement principle than of the wording. It is clear that once a condensation surface meets a regions with saturated vapors that condensation and growth leads to coating and incorporation of initially solid aerosols by/into stratospheric compounds such as H2SO3 - HNO3 - H2O. In the revised article inexact wording concerning this point is avoided after rephrasing.

Referee #3: For instance, the summary paragraph on Page 9867 (line 11-22) suggests that 'increasing particle mixing ratios as function of altitude inside the vortex are solely supplied by non-volatile particles' when it would be more realistic to say that they are primarily supplied by aerosol containing non-refractory material larger than 10 nm.

> **Author's reply to Comment:** After rephrasing, the paper now states at this point:
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> […] However, for the RECONCILE case this means:
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> (a.) Increasing particle mixing ratios as a function of altitude inside the vortex are primarily supplied by aerosol containing refractory cores. Otherwise, N_{10}-N_{10}nv would generally show a similar increase with altitude, and
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> (b.) If N_{10}-N_{10}nv includes non-volatile residuals too small to be detected ($d_p < 10$ nm), then the particles descending inside the vortex during RECONCILE consisted predominantly of non-volatile particles larger than 10 nm. [...]

Referee #3: There is ample evidence of this new particle formation in the 25 km range from the OPC and other data sets. Recently MIPAS measurements have shown that SO2 abundances have a strong gradient at this same altitude range (decreasing downward) supporting the idea that the aerosol is primarily sulfate (see papers by Hoepfner). Climate models using interact aerosol formation show a similar phenomenon. It is not inconceivable and in fact it is likely that the aerosol forms on down welling nuclei of some sort whether they are meteoritic or terrestrial in origin. This sulfate originates in the tropics where aerosol evaporates in the tropical stratospheric upwelling associated with the Brewer-Dobson circulation and which subsequently finds its way to high latitudes and then is caught up in the winter time down welling associated with the vortex. It should also be noted that the refractory portion of these aerosol will be transported along with gas phase sulfur (SO2 mostly) and likewise descend in the winter vortex where they can serve as nuclei for the

reforming sulfate aerosol. These could have a quite diverse composition including sea salt, surface material, soot, and even recycled meteoritic material.

> **Author's reply to Comment:** The comment touches one of the previous points (and comments by the other referees) that will be accounted for with the rephrasing in the revised version. We fully agree that the stratospheric aerosol may primarily consist of sulfate compounds and that refractory residuals may also have other origin than only the meteoritic ablation. Thus the refractory aerosol detected in the arctic vortex may indeed consist of material including sea salt, surface material, soot, and even recycled meteoritic material. The COPAS technique, this article is focusing at, does not allow for distinguishing the chemical species and thus does not give any hint about specific sources. As the meteoritic influx appears to be one of the most prominent sources due to the excessive amount of material that is imported into the atmosphere our estimate may serve to evaluate if this prominence is justifiable.

Referee #3: Page 9860; line 15-19; unless I just totally do not understand what the authors are doing here, this is a major error. I assume that the authors are using these size distributions as if they represent the size distribution of the refractory material, scale the number density and integrate to get a total volume and compute mass. However, all of these size distributions are for total aerosol size distributions that are primarily liquid sulfate aerosol which may/probably have solid, non-volatile inclusions within them. Any size distribution of these inclusions is likely to be largely uncorrelated to these size distributions and any refractory material masses inferred using these are almost certainly incorrect (and much too large). The authors must clarify how they are employing these size distributions and use realistic distributions or admit that they do not exist.

> **Author's reply to Comment:** In correspondence to the reply to Referee 1 and 4 as here the same subject is focused:
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> We reconsidered the various arguments and recalculated our estimates for the upper and lower limits.
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> **General approach:** We try to arrive at an estimation of the total mass of refractory particulate matter contained inside a Northern hemispheric winter vortex using (a) our measurements and (b) assumptions based on what is available in the literature. This necessarily implies large uncertainties and contains a certain level of speculation, still. As soon as better data or parameterizations become available in the future the numbers may change accordingly. However we think the approach in general is valid, if all the caveats are clearly pointed out. Along these lines we hope to have improved the manuscript after considering the reviewer's comment.
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> **(1.) Uppermost limit:** The previous calculation of the submitted manuscript serves now as an uppermost –theoretical- limit of our estimate. Yes, indeed these size distributions are provided for total stratospheric aerosol that assuming primarily liquid sulfate aerosol. If all this were refractory matter this would be the highest possible mass limit. The revised article version now also states that these size distributions are used due to the general lack of realistic, parameterized size distributions of the refractory portion of the stratospheric aerosol at this region in the atmosphere.
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> **(2.) Lowermost limit:** In the revised version now the numerically modelled size distribution of meteoritic ablation material without sulfuric acid cover is considered that is provided by Bardeen et al. 2008. This computed size distribution is given for 30 km as the lowermost altitude, thus, still somewhat above the highest level of our measurement. Descending, this size distribution may even shift further towards larger particles sizes between 30 km and 20 km altitude. As part of the above mentioned speculation we used this size distribution at 30 km altitude from Bardeen et al. (2008) as the extreme lower limit for our estimate.

(3.) Density differences: Moreover, our estimates are furthermore recalculated with an increased range of material densities (now 1000-3000 kg per m^3) as reasonably suggested by one referee. 2000 kg per m^3 is used as proxy for a mean material density.

Based on (1.) through (3.) renewed estimates resulted in changed values in the revised version. The estimate is limited at two sides:

A) the size distribution of refractory material cannot extend beyond the size distribution of the stratospheric sulfuric acid background aerosol, represented by the work of Jaenicke, Wang and Deshler.
B) the size distribution of refractory material may not undercut the modelled size distribution of meteoritic ablation material after transport from 90 km down to 30 km altitude (Bardeen et al., 2008).

As a matter of fact we are grateful to the reviewer's insistence on these points as the newer values now probably provide a much better estimation.

Referee #3: For that matter I am not totally convinced that an aerosol would necessarily have only one nuclei since they are the end result of coagulation of many smaller particles for which some number of them could include independent inclusions. Perhaps these stick to each other and form a more complex inclusion; I don't know. (relevant to Page 9876, line 3-6). Unless, I have totally misunderstood what is being done, the numbers discussed in detail on Page 9879, line 12 and further are not correct.

Author's reply to Comment: Correspondingly to the reply to a comment of Referee 2: The COPAS CPC technique unfortunately does not allow for quantifying the uncertainty of several particles incorporated in one sulfuric acid droplet. The technique is not able to distinguish whether one single core or a few nuclei are contained in one H2SO4 particle and if, in latter case, after vaporization of the volatiles a single remnant is counted or if the residuals re-separate into fractions (which seems not probable). For clarification we included at page 9860, at the end of the section starting with line 15:

[...] Note that an individual stratospheric sulfuric acid particle may incorporate more than one refractory core. The COPAS technique does not unambiguously allow for assorting an individual refractory residual to a single sulfuric acid droplet. It also does not allow for a strict conclusion as to whether multiple refractory incorporations adhere together after the volatile aerosol compounds are vaporized due to the heated COPAS aerosol line. We assume, however, that after contraction due to the surface tension of each evaporating droplet, the van-der-Waals forces will keep the remaining refractory residuals in shape of a single particle. [...]

Comments:

Referee #3: Page 9855, line 3-6; I don't see how the first half and second half of this sentence goes together. At the least, this statement cannot be made without further support.

Author's reply to Comment: The statement is erased in the revised version.

Referee #3: Page 9866, line 12-14; this is an over generalization and doesn't add thing to the text. I would remove it or add appropriate caveats.

Author's reply to Comment: Removed as suggested

Referee #3: Page 9869, line 3-5; Here the authors are neglecting other sources of non-volatile particles.

Author's reply to Comment: In the ACPD manuscript it is stated: "a strong presence of meteoric ablation materials can be assumed" which implies that other sources may contribute. However the section is rephrased into:

[...] For these reasons it can be assumed that the thermo-stable aerosols are predominantly comprised of meteoric ablation materials, although detailed chemical analyses of such particles in the submicron size range are still scarce (cf. Murphy et al., 2013 and references therein).[...]

Referee #3: Page 9876, line 18-20; A factor of 20 is a huge uncertainty and makes me wonder (beyond other strong trepidations about this analysis)

Author's reply to Comment: We agree with the referee about the scale of the uncertainties. Indeed the specified value was wrong due to a transcription error. The revised version contains the recalculated uncertainty factors and at this concrete point the factor is 5 instead of 20. Of course, this still is a large uncertainty. Nevertheless, we think that the estimate has its value as (a) we outline a new method for estimating the refractory matter, and (b) we specify how uncertain the values may be which are obtained from this estimate.

Minor comments:

Referee #3: Page 9852, line 5-7; I think this sentence is has too many superlatives particularly given the large uncertainties cited later in the paper.

Author's reply to Comment: Sentence is erased

Referee #3: Page 9853, line 3; 'is' not 'to be'

Author's reply to Comment: corrected as suggested

Referee #3: Page 9866, line 18-23; I don't follow this section at all.

Author's reply to Comment: Following also the recommendation of another referee this particular section, as a large fraction of the entire paper, is comprehensively rephrased. Please refer to the revised article.