Anonymous Referee #4

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 #4: […], whereas the presentation of the material is not acceptable for publication in its current form and requires substantial modifications before re-consideration. Major topics to be addressed before publication are the following:

1/ The style of writing requires major clarification because actually some paragraphs are simply not understandable. This point was raised by all other reviewers and there is no need to repeat their arguments here.

> **Author's reply to Comment:** Following the recommendation of most of the referees almost the entire article is restructured and rephrased.

Referee #4: 2/ The N2O tracer is used to create an index describing the air mass origin. Index values separating regions of different air mass origins are described and a reference is given where the vortex index is described. Since this index is key in the interpretation of data, it is not acceptable that the only reference to this index is cited as "in preparation". If the companion paper is not yet submitted and thus not accessible to the reader, some key information on the vortex index has to be given here, e.g. information on the justification of the index and its accuracy. How precise in the separation of air mass origin by this index?

> **Author's reply to Comment:** The respective paragraph has been rewritten to better explain and justify the empirical index, and a reference has been added to a published study (Greenblatt et al., 2002) upon which the idea of the index is based:
>
> […] Due to the strong subsidence and dynamical isolation of air inside the vortex, N_2O and other long-lived tracers exhibit sharp meridional gradients at the vortex edge. Greenblatt et al. (2002) demonstrated that the inner edge of the Arctic vortex can be accurately determined by the excess of measured $N_2O_{meas.}$ relative to characteristic values inside the vortex at a given potential temperature and at a given time, i.e. $\Delta N_2O = N_2O_{meas.} - N_2O_{vortex}(\Theta)$. At levels around 450 K this study (Greenblatt et al., 2002) found a ΔN_2O value of $\sim$ 20 nmol mol^{-1} to generally correspond well to the dynamical Nash-criterion (Nash et al., 1996) which is a commonly used criterion for defining the vortex edge. Extending this concept, we here define what we denote as the ΔN_2O vortex index (ξ_{vi}) as:
>
> $$\xi_{vi} = \frac{\Delta N_2O(\Theta)}{N_2O_{mid-lat.}(\Theta) - N_2O_{vortex}(\Theta)} \qquad (1)$$
>
> Essentially, the N_2O mixing ratios are linearly rescaled to altitude-independent characteristic index values of $\xi_{vi} = 1$ inside the vortex, and $\xi_{vi} = 0$ at mid-latitudes. This vortex index will be introduced and evaluated in detail in a further study (Hösen et al., 2014), which will also demonstrate that ξ_{vi} can, with some caveats, be viewed as an empirical proxy for the fraction of vortex air in an observed air mass. As the vortex index is conserved on time scales of isentropic transport, it essentially provides information on whether a measured air mass originally came from the interior of the vortex or from the outside. Therefore, the COPAS measurements can be categorized with respect to their recent origin by means of the index ξ_{vi}. The theoretically maximum value of ξ_{vi} is one, indicative of pristine vortex air mass. The criterion limits defined for this study to classify the COPAS measurements for Θ-levels above 400 K are:

1) $\xi_{vi} > 0.75$: Sample air originating primarily from the vortex interior,
2) $0.75 < \xi_{vi} < 0.25$: Sample air with extra-vortex, mid-latitude air contributions which are too large for unambiguous apportioning.
3) $\xi_{vi} < 0.25$: Sample air originating from well outside of the polar vortex, consisting mostly of mid-latitude air.

Since the ξ_{vi}-range between 0.4 and 0.8 is populated by relatively few air samples recently originating from the vortex edge region, the used criterion limits are regarded as sufficiently severe for categorizing the COPAS measurements.[…]

Referee #4: 3/ The deduction of total nv-aerosol mass is not clear. The authors used size distributions measured for the total stratospheric aerosol, including volatile particles and scaled these size distributions until the related number densities matched the observed values for nv aerosols. This approach assumes similar size distributions for nv and total aerosol which, however, is questionable. Furthermore they used a density of 1400 kg per m3 without further justification of explanation how they derived this value. A critical review of the method of estimating total mass of nv aerosol is strongly recommended.

Author's reply to Comment: In correspondence to the reply to Referee 1 and 3 as here the same subject is focused:

We reconsidered the various arguments and recalculated our estimates for the upper and lower limits.

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.

(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.

(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 #4: 4/ Section 4 on Observations and Results contains a huge amount of details in running text which makes it almost impossible to identify the key information. It is strongly recommended to rearrange the section and include tables which contain the data. Then the text can focus on the differences and similarities of the different missions and respective atmospheric conditions.

Author's reply to Comment: We agree with the referee that specified section needs rearrangement and rephrasing which is addressed with the revised version. Additional tables, instead, are avoided but descriptive text is minimized.

Referee #4: 5/ Section 5 is focusing more on implications of the observations than on a discussion of results. Renaming the section title is thus recommended. Furthermore, the entire first subsection 5.1 should be part of the introduction than of the discussion section. Only the every last sentence (page 9870, lines 9-13) of this subsection refers to the presented material. In addition Section 4.2 on particle mixing ratios as function of N2O mixing ratio and Section 5.1 on vertical profiles of nv-aerosol mixing ratios treat almost the same subject and should be combined.

Author's reply to Comment: We partly agree with the referee and changed the title of this Section 6 (previously Section 5) into" Implication of the observations and discussion" As part of the discussion in Section 6 our conclusions are evaluated in the context of current knowledge and much of these find support by other studies. We agree to move the very first paragraph of this section into an earlier section but prefer to keep the comparison with previous studies of aerosol detection in current Section 6.

Referee #4: 6/ Sub-section 5.3 on Implications for PSC formation requires more in-depth discussion. Although the authors observed almost 75% by number of the total aerosol being non-volatile they hypothesize only a small contribution of nv-aerosol cores to PSC formation. What is the reason for this discrepancy?

Author's reply to Comment: We added some more detailed explanation in this subsection, as suggested.

MINOR COMMENTS

Referee #4: 1/ Abstract, line 10-13: this sentence is not understandable, please rephrase.

Author's reply to Comment: rephrased into:

[...] Inside the vortex and at potential temperatures $\Theta \geq 450$ K around 11 submicron particles per cm^3 were generally detected. Up to 8 of these 11 particles per cm^3 were found to contain thermo-stable (at 250°C) residuals with diameters of 10 nm to about 1 µm. [...]

Referee #4: 2/ Page 9852, line 7: deposited "in" the upper atmosphere.

Author’s reply to Comment: corrected as suggested

<u>Referee #4:</u> 3/ Reference Murphy 2013: please check the reference in the reference list, obviously there is the wrong paper referenced.

Author’s reply to Comment: corrected

<u>Referee #4:</u> 4/ Page 9854, lines 26 – 29: this sentence is not understandable, please rephrase.

Author’s reply to Comment: rephrased into:

[…]It seems very plausible that such particles support other microphysical processes, such as heterogeneous freezing, although bulk freezing experiments (Biermann et al., 1996) indicate minor importance. […]

<u>Referee #4:</u> 5/ Page 9859, line 8: shouldn’t this be a “suit of inert artificial tracers”?

Author’s reply to Comment: the correct expression (used in numerous papers, e.g. Volk et al., Science 1996) is in fact “a suite of tracers”.

<u>Referee #4:</u> 6/ Page 9875, line 13: please rephrase “…the temperatures will become too high …”

Author’s reply to Comment: corrected as suggested

<u>Referee #4:</u> 7/ Check references in the text, very often brackets are missing for years of publication.

Author’s reply to Comment: corrected as suggested