

Anonymous Referee # 1:

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 # 1: 9853.15-21 – What do “those bodies” and “they” refer to? The text suggests particles less than a mm are not heated by friction, now here it seems to suggest they are, and that particles less than 10⁻⁵ kg are fully vaporized. I am confused.

Author’s reply to Comment: section is rephrased into:

[...]Furthermore, von Zahn (2005) estimated that objects in the mass range of 10⁻¹¹ to 10⁻⁵ kg correspondent to diameters of 20 μm - 2 mm contribute the major part of the total influx. These predominantly sub-millimeter sized particles are nearly fully vaporized mainly in the altitude range between 75 – 120 km [...]

Referee # 1: 9857.18 ... or from ...

Author’s reply to Comment: corrected as suggested

Referee # 1: 9865.4-5 ... are compared in Fig. 5 in ... The authors should mention that in many cases the percentile bars do not exceed the data points.

Author’s reply to Comment: text is corrected as suggested and comment concerning percentiles is inserted in the caption:

[...] If whiskers are not visible the percentile range is covered by the size of data points. [...]

Referee # 1: 9866.21 The 75th percentile just barely exceeds 75% if you look really closely. This sentence is a stretch and not that important so should be deleted.

Author’s reply to Comment: corrected as suggested

Referee # 1: 9866.25-9867.2 Very awkward language. Try the following. The contribution of volatile and semi volatile particles is accounted for with the mixing ratio difference given as $N_{10} - N_{10nv}$. Coated non-volatile particles greater than 10 nm will be included in N_{10} , but once their coating is removed they will not be sensed in N_{10nv} . Thus this measurement cannot tell the difference between volatile and partly volatile particles which are near 10 nm.

Author’s reply to Comment: Following the recommendation of the referee text is rephrased into:

[...] With the mixing ratio difference given as $N_{10} - N_{10nv}$ the contribution of volatile and semi-volatile particles is accounted for. As long as coated non-volatile particles have diameters larger than 10 nm they are included in N_{10} . Once the volatile coating is removed, releasing a residual with $d_p < 10$ nm, this remnant is not accounted for with N_{10nv} . Therefore, the COPAS measurement technique does not allow for distinguishing between either a fully volatile particle or a semi-volatile particle with a diameter of close to 10 nm. [...]

Referee # 1: 9868.1-16 This is an odd paragraph. The first sentence provides the obvious general observation most important for this paper that N_{10nv} and f increase with decreasing N_{2O} and thus increasing altitude. Then the discussion starts at the base of the profile, near the tropopause, and works its way up until the first sentence is essentially repeated. Do we

need all this intermediate discussion? If so make a choice and start from the bottom and arrive at the top, but don't repeat the point. Also while it may be obvious to the authors that these high values of N_{10nv} near the top indicate downward transport, this has not shown to the reader up to now, and thus is speculation. It should be indicated as such, or proven, or left out here.

Author's reply to Comment: The first two sentences of this paragraph were erased. We follow the idea to start from the bottom and arrive at the top and leaving one conclusive statement afterwards.

Referee # 1: 9869.3 Do the authors mean above 250 C?

Author's reply to Comment: the section is rephrased into:

[...] In summary, the observations reveal that inside the polar vortex and above 490 K potential temperature up to $150 \text{ mg}^{-1} (N_{10nv})$ from a total of $200 \text{ mg}^{-1} (N_{10})$ are thermally stable at 250°C . Therefore, a large fraction of investigated aerosols does not evaporate at 250°C and contains materials other than H_2SO_4 , HNO_3 , H_2O or other volatile compounds. [...]

Referee # 1: 9869.5 Scarce or non-existent? There is a difference. Have such particles ever been chemically analyzed?

Author's reply to Comment: "scarce" is meant, indeed: There are real-time chemical analyses of particles which are assumed to be of meteoritic origin provided by the group of D. Murphy (NOAA, Boulder, US) and published for instance by Cziczo, Murphy and Froyd from mid-latitude UT/LS measurements – which is at least more than "not-existent".

The reference to Murphy et al., 2013 is now added at the end of this section.

Referee # 1: 9869.22 January of the year 1990 or January 1990? If I remember correctly these measurements by Hofmann et al. were at much higher altitude than the observations here and by Wilson et al.

Author's reply to Comment: 9869.16-22 rephrased into:

[...] The study of Wilson et al. (1990) already described increasing submicron aerosol concentrations at altitudes of up to 20 km inside the Arctic vortex by measurements utilizing the NASA *ER-2* aircraft during the Airborne Arctic Stratospheric Expedition (AASE) in January and February 1989. Further evidence for an increasing number concentration of condensation nuclei (CN) as a function of altitude was found from balloon-borne measurements with significant excess at higher altitudes (between 20 - 28 km) at Kiruna in January 1990 (Hofmann et al., 1990, Figure 1a therein). [...]

Referee # 1: 9871.6 What has a tropospheric origin, the particles or the sink?

Author's reply to Comment: This sentence does not exist anymore in the revised paper.

Referee # 1: 9871.26-9872.2 The parenthetical statement is confusing and unnecessary. The last sentence is very awkward.

Author's reply to Comment: The whole section is rephrased – please see next comment:

Referee # 1: 9870.21 – 9871.9 This section is hard to follow. The justification for the imaginary grey curves is not well explained. There are no data to support the grey lines, and 5 separate considerations, used to establish the lines, are too many to be fully satisfied or understood by the reader. Plus the conclusions from this section are not necessary for the overall result of the paper. Remove this discussion and the grey lines in Fig. 6.

Author's reply to Comment: We restructured and simplified the respective section, in particular the discussion and detailed hypothetical reconstruction of the (not observed) canonical correlation. Instead we added text emphasizing that the evolution of the correlations and profiles suggests significant downward transport of particles by diabatic dispersion within the vortex. We believe this discussion is important (and should certainly not be removed as the reviewer suggests) as it identifies a major transport process into the lower stratosphere. Figure 6 is modified: the remaining grey lines in this Figure 6 provide the congruent of the ESSenCe correlation – as a kind of reference for the late winter correlations. Still the grey lines depict an extrapolated progress for altitudes above the observed levels. Further lines and grey shaded areas as given in the earlier version of Figure 6 are erased.

[...] The observed correlations between N_{10nv} and the long-lived tracer N_2O can be consistently interpreted in terms of the theory of stratospheric tracer-tracer correlations which is well developed and verified by observations (cf. Plumb, 2007, and references therein).

In the absence of the polar vortex, rapid isentropic mixing creates a unique extra-tropical canonical correlation between two long-lived tracers. The shape of the canonical correlation of tracers is determined by the vertical distribution of the respective sources and sinks. In particular, this canonical correlation is expected to exhibit curvature in the region close to sinks or to sources of either compound, but to be linear elsewhere.

After the formation and ensuing subsidence of the polar vortex the polar transport barrier isolates the air inside the vortex. As a consequence the correlation within the vortex may change over the course of the winter due to diabatic dispersion within the vortex and/or in-mixing of mid-latitude air. For reasons explained in Plumb (2007) the effect of these processes is a progressive straightening of the correlations. Thus, while the curved canonical correlation is expected to remain almost unchanged at mid-latitudes, the correlation inside the vortex deviates from the canonical shape due to the vortex driven downward transport of refractory aerosol originating from a source at high altitudes.

The grey lines in Figure 6 are congruent with the ESSenCE correlation inside the vortex, qualitatively extrapolated by its expected continuation toward lower N_2O values. Above the sampled altitudes N_2O continues to decline and eventually converges towards zero in the mesosphere while N_{10nv} will further increase by approaching the source region of the refractory aerosol.

Note that the correlations cannot change due to the mean large-scale subsidence. The correlation could only deviate from canonical shape due to diabatic dispersion and/or in-mixing from mid-latitudes (cf. Plumb, 2007). However, these processes, dispersion or in-mixing, would have different effects on the evolution of the vertical profiles. (1) Mid-latitude in-mixing would tend to decrease particle mixing ratios at a given potential temperature above 410 K, thus counteracting the mean subsidence. (2) Alternatively, diabatic dispersion would lead to additional dispersive downward transport of particles.

Because of the observed strong particle increase at all potential temperatures above 410 K between early and late winter (despite slow diabatic subsidence at these altitudes), we hypothesize that:

- 1) The diabatic dispersion is the dominant factor in the evolution of the correlations and likely also contributes significantly to the evolution of the vertical profiles.
- 2) The diabatic dispersion is thus becoming an important mechanism for the transport of refractory particles to the vortex bottom. [...]

Referee # 1: 9875.5 “Particle volume ratios per air mass”. What does ratios mean here? Isn't the calculation, the particle volume per mass of air, or aerosol volume mixing ratio?

Author's reply to Comment: It is indeed the total particle volume per mass of air.

Following the suggestion of other referees, the description of certain steps of our calculation was rephrased and is in general better understandable in the revised version.

Referee # 1: 9875.11 The authors should work harder on their English to make it more concise and simpler and thus more easily understandable. For example, “Total particle volumes and total particles masses (under consideration of a range of densities for the particulate material ...,” could be restated, Total particle volumes and masses for a range of particle densities.

Author's reply to Comment: See comment above

Referee # 1: 9878.7 Don't the authors mean, mesospheric air down to levels of 500 K?

Author's reply to Comment: rephrased as suggested

Referee # 1: 9878.11 Here and elsewhere what is meant by “vertical dispersion”? What is causing this? What is the direction of motion and how is it different than the vortex subsidence?

Author's reply to Comment: In correspondence to the reply to a comment of Referee 2:

for clarification text is inserted in section 1 and according discussion throughout the entire article is comprehensively revised:

[...] According to model studies of vertical vortex transport (e.g. Plumb et al., 2002) most of the content of the mesosphere is ingested by the vortex towards the end of a polar winter and the signatures of mesospheric air are discernible down to the vortex bottom. Balloon-borne observations of carbon monoxide (CO) have traced mesospheric origin down to levels of 500 K of potential temperature (see Figure 6 in Plumb et al., 2002), which is above the maximum ceiling of the M-55 *Geophysica*. The mean large-scale subsidence inside the vortex apparently occurs most efficiently at altitudes above 500 K. With subsidence rates of 1-1.5 K of potential temperature per day, as observed throughout the three missions EUPLEX, RECONCILE and ESSenCE, a transport further down to 400 K would exceed a period of three months. As a consequence, the vertical transport of refractory aerosol below 500 K of potential temperature may be mainly driven by diabatic dispersion inside the vortex rather than by the mean large-scale subsidence. Diabatic dispersion may be understood as a gradual vertical mixing down to the vortex bottom. This process has been found to be consistent with the development of observed tracer distributions inside the Arctic vortex (Ray et al., 2002).[...]

Referee # 1: 9879.1 Is the air mass really twice as large between 67 and 1 hPa compared to 100 to 67 hPa. This surprises me, although I didn't do the calculation, which is straight forward, so why do the authors state considering? This sounds like an assumption.

Author's reply to Comment: We fully agree with the referee, the calculation is straight forward: from integrating the hydrostatic equilibrium equation for the altitude dependence of the ambient pressure it follows that increasing the pressure difference by a factor of 2 results in the doubling of the air mass.

However, the sentence is rephrased into:

[...] since the air mass is twice as large in the column 67 to 1 hPa as in the observed column 100 to 67 hPa, we estimate [...]

Referee # 1: 9878.26 – 9879.11 Here is another example of the convoluted, and unnecessarily complicated language used by the authors. I provide my last example of how this could be rewritten so that the reader does not have to read and re-read the whole section to understand it.

Assuming the simulation of Plumb et al., 2002 to be realistic, and the fact that the air mass between 67 and 1 hPa is twice the air mass between 100 and 67 hPa, we estimate that at the end of the Arctic winter about 10–30% of the mesospheric air mass contribution to the whole vortex volume resides in the measurement region below 470 K (see Fig. 6 in Plumb et al., 2002). Assuming that the increase in the observed particle mass between 100 and 67 hPa from mid December to late winter, 32_106 kg for RECONCILE, can be attributed to fresh mesospheric particles, and that the outflow of these particles at the vortex bottom is negligible, compared to the import from aloft, leads to a mesospheric particle influx over the whole vortex of 107- 320 _106 kg for the RECONCILE winter 2009/2010. The reader can then easily see where the numbers come from and this doesn't have to be spelled out in a parenthetical comment.

Author's reply to Comment: Following the recommendation of the referee, the section is comprehensively rephrased.

Referee # 1: 9879.12 “ranges at about” but no range is given. Thus, “is about” makes sense, ranges about does not.

Author's reply to Comment: corrected as suggested

Referee # 1: 9879.13-14. deposited? Do you mean deposited here and elsewhere? Where does the 40e6 come from? I assume it is 110e3 per day times a fraction of the year. But why not use 1.1e5 kg/day and 4e7 kg? Then it is easy to see that 4e7 = 1.1e5*365?

Author's reply to Comment: “deposited” replaced by “deposited”
And indeed: 40×10^6 kg per year = 110×10^3 kg per day times 365 days. Text is rephrased into:

[...] The expected global influx of meteoritic material is about 110×10^3 kg per day (Love and Brownlee, 1993). Thus, per year up to 40×10^6 kg of meteoritic material may be deposited in the mesosphere. [...].

Referee # 1: 9879.19-23 Try. ... This discrepancy is within the range of our estimate ... the minimum of the uncertainties (cf. ...ESSenC). In addition the remnants ...

Author's reply to Comment: most of the specified text fragments are eliminated due to rephrasing and restructuring of the article.

Referee # 1: 9879.25 Try : ...Finally parts of the refractory aerosol in the Arctic vortex may originate from sources ... mesosphere. Thus our ... provide a highly uncertain upper limit, as our assumption is the observed refractory matter is solely of ...

Author's reply to Comment: see previous comment.

Referee # 1: 9880.8 The authors should be a bit more careful in separating the increases discussed by Wilson et al. near 20 km, and those discussed by Hofmann et al., which are quite a bit higher and the character of the increase is significantly different. The former a steady increase with altitude, while the latter a sudden rapid increase in a layer which then relaxes again. In addition the authors may want to mention the recent work of Campbell and Deshler, 2014.

Author's reply to Comment: Although the text in Wilson et al. 1990 speaks about 19.5 km maximum flight altitude the graphs in this paper show measurement points at up to 21 km altitude. Further above, Wilson et al. was not able to measure due to the limited maximum flight altitude of the ER-2. Wilson et al. provide CN number concentration with logarithmic scale running from 4 cm⁻³ to 20 cm⁻³. From the study of Wilson et al. the averaged number concentration increases either from about 5 to 7 cm⁻³ or from slightly below 10 to 11 cm⁻³ between 19 and 21km altitude.

The Hofmann et al. 1990 balloon-borne measurements allowed for measuring at up to 28 km. Essentially Hofmann et al. focus at NAT formation in their paper, at levels clearly above 20 km. Nevertheless, the graph in Hoffmann et al. 1990 (Fig1a), with a logarithmic scale of number concentration in cm⁻³, that covers 7 orders of magnitude, shows a turn of the vertical progress of the CN profile at about 20 km altitude with increasing slope further above. Probably it is over-interpreted, but this turn in the vertical progress agrees qualitatively and quantitatively pretty well with the observation of Wilson et al. 1990.

Despite the problem that a reader has to guess if the altitude ranges of the two independent measurements have an overlap above 20 km or not, it seems plausible that both authors describe the same subject. Wilson et al. probably observed the base level of this phenomenon that they interpreted to be caused by new particle formation. Instead, Hofmann et al. 1990 had the opportunity to provide a view on likely the same phenomenon to much higher altitudes. However, though not further investigated by Hofmann et al. (as not a subject of their paper), Hofmann et al.'s CN measurements (Fig1a) at the same place and time as those of Wilson et al. strongly support the observations by Wilson et al.

The work of Campbell and Deshler, 2014 is now cited in revised manuscript version.

Referee # 1: 9880.22 What does the etc. refer to?

Author's reply to Comment: the "etc" refers to other sources but for clarity is replaced by "or other sources"