

Anonymous Referee # 2:

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 # 2:

[...] but there are several issues which need to be addressed first.

The first concerns the nature of the flux of interplanetary dust particles (IDPs), and the formation and transport of MSPs. Most of the mass entering the atmosphere is carried by particles around 10 microgram in mass (100 micron in radius). The bulk of these particles ablate, producing a vapour of metal atoms. Over several days, this vapour is oxidized and polymerises to form MSPs, initially around 1 nm in radius (these are the particles measured by rocket-borne detectors). IDPs smaller than about 0.1 microgram (20 micron radius) do not ablate, and these particles sediment rapidly into the troposphere (the statement at the top of page 9854 is incorrect). In contrast, MSPs are too small to sediment, and are transported by the residual circulation. These particles descend in the polar vortex over several months, during which time they coagulate to become larger than 10 nm. In fact, it takes about 5 years for these particles to reach the Earth's surface (Dhomse et al., GRL 2013). This is because of the nature of the meridional circulation, which causes the particles to "wash" back and forth between the polar vortices. Eventually, they escape into the lower mid-latitude stratosphere, and then enter the troposphere. These processes have been modelled using 2D (Megner et al., JGR 2008) and 3D (Bardeen et al., JGR 2008; Saunders et al., ACP 2012; Dhomse et al., 2013) models. None of these studies are cited in the present paper, though they are central to interpreting the measurements.

Author's reply to Comment: We agree with the referee that the manuscript is lacking of suggested literature references. The updated manuscript version considers specified references and involves the results of these, and further studies in the discussion. Nevertheless, we do not see any incorrectness in the statements on page 9854. As the meteoric influx is supposedly a constant process, the sedimentation of the larger (IDP) particles through the region of our measurement should also be a continuous process. Thus, we can expect particles in the probed air between the minimum size determined by condensation and coagulation and the maximum size determined by the 'non-ablation'.

Please refer to the rephrased Section 1 in the revised version wherein the specified studies are included and the text is rephrased to clarify the difference between IDPs and MSPs and their contribution to the refractory aerosol.

Referee # 2: Another point that needs to be made in the paper is that small MSPs can accumulate in sulphate particles (and PSCs) in the lower stratosphere. Thus the technique used here, which drives off the volatiles to leave a refractory residual, measures the sum of MSPs that have accumulated in each stratospheric aerosol, which is therefore not a measure of the size distribution of MSPs entering the lower vortex from the mesosphere.

Author's reply to Comment: We partly agree with the referee. The COPAS CPC technique unfortunately does not allow for quantifying the uncertainty of multi-MSP-incorporation into 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 following text:

[...] 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. [...]

The counted refractory aerosol particles of course may not represent the size distribution of MSPs subsiding from aloft. But to cover a wider range of uncertainty the underlying range of size distributions for our estimate is expanded to one size distribution provided by Bardeen et al., 2008, for altitudes of 30 km over the winter pole. This will serve as the lowermost limit of our estimate of the total aerosol mass in the revised manuscript version.

Referee # 2: p. 9870, line 8. Refer to the modelling studies above to give more detail to this statement about the range of particle sizes observed.

Author's reply to Comment: Following text (and references) added for clarification:

[...] Of course, it cannot be ruled out that many refractory residuals were too small (if $d_p < 10$ nm) to be detected with COPAS. Such ultrafine particles are produced by re-condensation of metallic vapors from meteoritic ablation to form MSPs (Megner et al. 2008; Saunders et al., 2012; Dohmse et al., 2013). Particles in this size range, for instance if involved in noctilucent cloud formation, could be transported down to the middle stratosphere. Plane, (2012) suggests by referring to numerical studies (Bardeen et al., 2008; and Megner et al., 2008) that these ultrafine particles most likely agglomerate to diameters of up to 80 nm, e.g. driven by electrical charges, before entering the middle stratosphere from above.[...]

Referee # 2: p. 9876. Step 3 of this procedure is unclear. What does a “range of mean particle volumes ...” actually mean? Is this as a function of height? It sounds dangerous to take means of volumes because of the non-linearity involved in the volume distribution of a population of aerosols.

Author's reply to Comment: “Range” should refer to range of uncertainty, which is considered as the maximum variability of the available size distributions, expressed as average particle volume. Non-linearity was accounted for. As our wording was obviously misleading, we have rephrased the steps of calculation. See Section 6.4.1 in the the revised manuscript.

Referee # 2: p. 9877. Statement (d) needs some references to the “literature”

Author's reply to Comment: Following the referee's advice the statement (d) is modified into

[...] (d.) Due to the COPAS activation limit and the inlet transmission, particles of diameters smaller than 10 nm and larger than about 1 μm are disregarded (Weigel et al., 2009) [...]

Referee # 2: p. 9879, lines 12 - 29. This is a critical part of the discussion, yet is very brief. The estimated global input rate is enormous - see the review by Plane (Chem. Soc. Rev. 2012) which shows that the existing range of estimates is 3 - 300 t d⁻¹, whereas the estimate proposed here is 550 - 1700 t d⁻¹ !! If the authors examine the modelling studies above, they will see that MSPs spend a much longer time in the middle atmosphere than the assumption that the measurements here represent a single season's input. There is also a very brief statement about non-cosmic refractory particles contributing - is there something more to say about that?

Author's reply to Comment: In the revised paper the mass estimate is recalculated by using a size distribution provided by Bardeen et al., 2008 as a lowermost limit. Of course, it was not meant that a single seasons input is flushed via the vortex within the consecutive Arctic winter. Nevertheless, to keep the mass balance constant, the amount entering the mesosphere from above should somehow equal the amount that exits the mesosphere towards the surface. Otherwise particles would accumulate in the mesosphere. The COPAS measurement itself unfortunately does not allow for apportioning the refractory material to its origin. So a statement concerning possible contributions other than the cosmic input is not possible based on COPAS data only.

For clarification following statement is inserted in the revised paper version:

[...] Parts of the recently deposited aerosol material may remain in the mesosphere for several years (Dhomse et al., 2013). Nevertheless, a certain fraction of the recently deposited material may descend out of the mesosphere due to the vortex-induced subsidence during the next polar winter. When the vortex disintegrates in early spring the particles are horizontally spread towards mid-latitudes over the entire vertical extension of the former vortex column. Over the following seasons, until a new vortex can form, a certain fraction of particles may remain in the region above the pole. This fraction is available for incorporation into the newly forming vortex leading to further descent of particles. Finally, these particles reach the lowermost part of the vortex at $\Theta < 500$ K ahead of the newly incoming mesospheric air in early winter. However, for a balanced mass budget, the amount of material exiting the mesosphere towards the stratosphere should be in the range of the mesospheric input. Otherwise the meteoritic ablation material would accumulate in the mesosphere.[...]

Minor points

Referee # 2: The authors refer to micrometeorites (e.g. line 25 on page 9852) entering the atmosphere. In fact, (micro)meteorites are interplanetary dust particles which survive entry and eventually reach the earth's surface intact. The term used to describe IDPs entering the atmosphere is meteoroids.

Author's reply to Comment: corrected as suggested

Referee # 2: p.9857, line 8: N₂O is actually produced in the mesosphere by energetic particle precipitation, so this sentence needs to be rephrased. The mesospheric source is probably minor, so the interpretation in the paper of low-N₂O air coming from the mesosphere should be fine.

Author's reply to Comment: specified paragraph is rephrased:

[...] N₂O is generated at the surface and has its sink at high altitudes, generally above the tropopause, where with increasing altitude the N₂O molecules are destroyed by UV-photo-dissociation and reaction with O (¹D), oxygen atoms in an excited singlet state. Satellite observations of episodic N₂O enhancements in the polar mesosphere (Funke et al., 2008) also suggest the presence of a minor mesospheric source. Nevertheless, from a stratospheric perspective, air masses with low N₂O mixing ratio generally originate from high altitude, i.e. in the mid to upper stratosphere or mesosphere. [...]

Referee # 2: p. 9859, line 20: "led", not "lead"

Author's reply to Comment: corrected as suggested

Referee # 2: p. 9867, line 26: "monotonically", not "monotonously"

Author's reply to Comment: corrected as suggested

Referee # 2: p. 9871, line 10: how were the grey lines in Fig. 6 calculated?

Author's reply to Comment: Correspondingly to the reply to the suggestion of Referee 1 as here the same subject is focused:

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.

[...] 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 # 2: p. 9874, line 22: explain what "vertical dispersion" means.

Author's reply to Comment: In correspondence to the reply to a comment of Referee 1: for clarification new text is inserted in Section 1 of the revised version and according discussion throughout the entire article is comprehensively revised:

[...] 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 # 2: p. 9876, line 22: "particle's density"

Author's reply to Comment: corrected into "particle's material density"