Manuscript title: Enhancements of the refractory submicron aerosol fraction in the Arctic polar vortex: feature or exception?

**Autors general reply**: We gratefully acknowledge the repeated effort and engagement of the referees and the editor to improve this publication. In the following we provide the replies to the specific suggestions of one of the referees and changed the paper accordingly. For traceability of changes and modifications **any new text and adds to the previous paper version are highlighted in yellow**.

**Referee Comment:** Line 43, 76-78 and 807: The paper refers to the recent review by Plane (2012) which suggests that the meteoric input is in the region 10-100 t d-1, so why then state that 110 t d-1 is the "expected" value in the abstract and elsewhere? The Love and Brownlee estimate was obtained by assuming a velocity distribution for the particles hitting an impact detector, and the particle masses are very sensitive to the assumed distribution. If the mean velocity is shifted from 18 to 30 km s-1, the mass input would drop by an order of magnitude (see work by Janches and Mathews). The abstract should state the input estimated in the present study.

> **Autors reply**: The abstract now contains the estimated input based on our study. Sentence starting at line 40 now reads as:
>
> "Based on the derived increase of particle mass in the lower stratospheric vortex (100 – 67 hPa pressure altitude) by a factor of 4.5 between early and late winter, we estimate the total mass of mesospheric particles deposited over the winter 2009/10 in the entire Arctic vortex to range between 77 × $10^3$ and 375 × $10^6$ kg."
>
> In Section 1.1 furthermore following text is added:
>
> "…Plane (2012) presents a detailed discussion of these fluxes concerning plausibility and consistence with observations, and he limits the uncertainty in the flux estimates provided in the literature to a factor of 10 in the order of magnitude between ~ 10 to 100 × $10^3$ kg per day. The review by Plane (2012) clearly shows how uncertain the knowledge of the true meteoritic influx is. The value provided by Love and Brownlee (1993) was obtained by assuming a certain velocity distribution for the particles hitting an impact detector. The study of Janches et al. (2000) investigated the effective mass loss of meteoroids due to deceleration dependent on a broader range of penetration velocities, which potentially yields a refinement of the results of Love and Brownlee (1993). Nevertheless, we will refer here to the most accepted value of 110 × $10^3$ kg per day for comparing the results of our estimates, while bearing in mind that an uncertainty of one order of magnitude needs to be considered for the value of the meteoritic influx into the atmosphere (Plane 2012)."

**Referee Comment:** Line 66 and 81: inconsistent size limits for IDPs – which, by the way, are somewhat artificial definitions.

> ➔ **Autors reply**: the definition of IDPs in line 66 is adopted from specified reference.
> To avoid inconsistency the sentence starting in line 81 is rephrased into:
>
> "Such small objects are not sufficiently heated by friction with atmospheric air molecules when entering the atmosphere."

**Referee Comment:** Line 99: the AIDA chamber study described in Saunders et al (2010) did not look at H2SO4 uptake.

**Autors reply:** Indeed at this point the text was wrong. The sentence should read as follows and is now rephrased accordingly:

"Laboratory as well as modelling studies particularly investigate the potential of MSPs to act as condensation surfaces for water vapor ($H_2O$) to form ice clouds (Saunders et al., 2010) and their impact on stratospheric $H_2SO_4$ processing on global scales (Saunders et al., 2012)."

**Referee Comment:** Line 139: Section 1.2, not 1.3

➔ **Autors reply:** corrected into "1.2" as suggested

**Referee Comment:** Line 147: EEPs cover the precipitation of both electrons and protons – so SPEs are a subset of EEPs.

→**Autors reply:** the fraction "or solar proton events (SPEs)" is erased from the sentence.

**Referee Comment:** Line 225: EUPLEX has been mentioned in the abstract but not defined in the text before this point. Since the campaigns are defined in Section 3, each mention prior to that point should include a reference to the appropriate subsection of Section 3.

➔ **Autors reply:** a text reference is added to campaign names that appear prior to section 3.

**Referee Comment:** Line 295: add a reference for ERA-Interim

➔ **Autors reply:** The reference to Dee et al., (2011) is added at this point in the revised version.

**Referee Comment:** Line 404: this sentence is confusing. The variability of N10 particle densities is larger below 440 K than the slight decrease, which is only discernible because the variability is smaller above 440 K.

➔ **Autors reply:** the sentences is rephrased into:

"For $\Theta > 400$ K the mixing ratio $N_{10}$ remains fairly variable between 150 - 300 $mg^{-1}$ and becomes more compact with increasing $\Theta$ until 440 K with ~ 200 $mg^{-1}$ (Figure 2a and d)."

**Referee Comment:** Line 455: Is it "30-50" or "30-60"?

➔ **Autors reply:** we agree with the referee and the number is corrected into "30-60 $mg^{-1}$" in the revised version.

**Referee Comment:** Line 677: the statement "where the relative isolation of the vortex supports chemical reactions" requires explanation. As it stands, it makes no sense!

➔ **Autors reply:** the sentence is rephrased, 1) to avoid the conflict between "import of partices" from aloft and the "relative isolation", latter was related to isentropic in-mixing from lower latitudes. 2) heterogeneous chemical reactions, e.g. such as chlorine activations, are meant. These reactions likely are promoted by the presence of reaction surfaces provided by the imported particles:

"The import of refractory material into the vortex constitutes an important source of particles for a region where the isolation of the vortex from isentropic in-mixing promotes heterogeneous chemical reactions connected to ozone depletion."

**Referee Comment:** Line 684: replace "eminently" with "particularly" or "especially"

➔ **Autors reply:** corrected as suggested, replaced by "particularly".

**Referee Comment:** Line 728: using the size distributions of stratospheric aerosol does certainly provide an upper limit to the meteoric mass. But surely this upper limit can be reduced by having some idea of the volume of volatile to non-volatile material in the aerosol? Even if this is not known from the measurements, a sentence stating that this is the case would be helpful here.

- ➔ **Autors reply:** We fully agree with the referee. Therefore, text in the revised paper now reads as (end of Section 6.4.1):

  "(a.) About a factor of 5 in uncertainty is implied in $v_p$ from the range of size distributions. The therein included, but most important uncertainty arises from the unknown true size distribution of refractory particles with diameters between 10 nm - 1 μm. COPAS detects the particles of this size range, but does not size them. The use of the stratospheric aerosol size distributions does only provide an uppermost extreme for estimating the meteoric aerosol mass. The knowledge of the volume concentration of volatile in relation to non-volatile aerosol material would significantly reduce the uncertainty our approach is bearing. "

**Referee Comment:** Line 765: how do you know the VCSA uncertainty is up to 50%, is this just a guess?

- ➔ **Autors reply:** Since VCSA is computed from distributions of tracers indicating the origins of air masses, several uncertainties come together. First, the initialization depends on the computation of the vortex edge following Nash et al. (1996). While this method shows good results in regions where the horizontal PV gradient is steep, the assignment of the PV value denoting the edge in areas of flat PV distribution on the same isentropic level is somewhat arbitrary. This may lead to an inexact initialization already. Furthermore, during the course of the simulation the mixing parameterization leads to mixing in areas of strong deformation, leading to dilution of the vortex area (which is realistic). The mixing parameterization was carried out with values derived from Konopka et al. (2003). Although these values work sufficiently well for Arctic and Antarctic vortices, there may be dynamical situations when the model under- or overestimates the real mixing across the vortex edge. Taking into account sensitivity studies focusing on the above mentioned points we estimate the uncertainties in VCSA very conservatively to be less than 50%.

  For clarification specified sentence now reads as (end of Section 6.4.1):

  "(e.) The model estimates of the VCSAs also involve uncertainties of at the most 50 % as a conservative estimate of this uncertainty inherent with the accuracy of the computed vortex edge and of the simulation of the mixing parameterization."

**Referee Comment:** Line 979: "entire"

- ➔ **Autors reply:** In line 798 the error in writing "entire" was corrected

**Referee Comment:** Line 778-779: – Not sure what is meant by this sentence. What does "ranges" mean?

- ➔ **Autors reply:** sentence is rephrased into ("range" is erased)**:**

  "The mean particle volume derived from the MSP size distribution (Bardeen et al., 2008) is three orders of magnitude below corresponding values derived from the upper limit size distributions."

**Referee Comment:** Line 818-823: it is of course the case that the rate of deposition of meteoritic debris at the surface must equal the input at the top of the atmosphere. However, according to the 3D models the amount of material that descends in the polar vortex is not all removed at the end of the winter. Some of it is transported back into the mesosphere by the reverse circulation during the summer, and then descends in the vortex in the opposite hemisphere. This can happen several times, which explains why the average time the material spends in the atmosphere is 4 – 5 years (as referenced in the paper which refers to the study of Dhomse et al. (2013). Thus, the accumulation at the base of the vortex during one winter, which is used to obtain the meteoric input, is actually an upper limit.

- ➔ **Autors reply:** we fully agree with the referee that our estimate is highly uncertain, and that the estimated total aerosol mass within the vortex provides the uppermost extreme. This is stated several times throughout the paper.

**Referee Comment:** Line 832 – 'if adjusted for a size range of dp >10 nm' is the total mass used here the sum of only the particles > 10 nm in the Bardeen size distribution? Elsewhere in the current paper it is argued that particles would agglomerate on evapouration of liquid droplets, such that particles which Bardeen simulated as smaller than 10 nm would be detected by COPAS. This point needs to be clarified.

- ➔ **Autors reply:** Indeed, the total mass derived from the Bardeen size distribution is the sum of particles > 10 nm as this is exactly the particle size range detected with COPAS. Indeed it is argued in the paper that an evaporating droplet likely releases one remnant that is an agglomeration of initially individual particles. But is very unlikely that only sub-10nm particles are selectively activated and contained in one stratospheric aerosol droplet that, after evaporation, releases one COPAS detectable remnant. Instead sub-10nm particles may be incorporated in droplets that already contain refractory cores of larger sizes. Condensation happens preferably onto larger condensation nuclei within a given size distribution due to the smaller contact angle that is provided to the condensing medium. Furthermore, the mass contribution of particles with sizes < 10nm is negligible compared to the integrated mass of all particles from the size distribution of Bardeen et al. (2008) with size > 10 nm.

  For clarification, however, one sentence is added:

  "The size distribution of Bardeen et al. (2008), if adjusted for the size range of $d_p$ > 10 nm to be in accordance with $N_{10}$nv, results in an in-vortex mass of refractory aerosol that is much smaller than could be expected. The resulting refractory aerosol mass from the size distribution of Bardeen et al. (2008) does not consider particles of sizes $d_p$ < 10 nm as these particles are not detected by the $N_{10}$nv channel of COPAS and as their mass contribution is negligible."

**Referee Comment:** Line 837: maximum, not minimum?

- ➔ **Autors reply:** sentence is rephrased into ("minimum" is erased)

  "The true refractory aerosol size distribution, which is currently unknown, is very likely located somewhere in between the stratospheric background aerosol (Jaenicke, 1980; Wang et al., 1989; Deshler, 2008) and the numerically derived size distribution of MSPs (Bardeen et al., 2008)."

**Referee Comment:** Line 841-843: see earlier comment about the size distribution of the refractory material being shifted to significantly smaller size than the stratospheric sulphate aerosol. Confusingly, this is recognised in lines 851-852!

➔ **Autors reply:** the section is re-organized. The sentence in lines 851-852 is erased and the last sentence of this paragraph was moved behind the sentences in line 841-843:

"Thus, it seems conceivable that the true refractory aerosol size distribution is very similar in shape to the size distribution of the stratospheric background aerosol, but this true refractory aerosol size distribution may peak at a certain smaller particle size. As a hypothesis, we assume here that the distribution peaks closer to our estimate's upper limit rather than being strongly shifted towards the estimate's lowermost limit. To sufficiently drain the expected meteoritic influx the true size distribution should result in an integrated refractory aerosol mass inside the entire vortex that is of the magnitude of the half-year influx, i.e. $20 \times 10^6$ kg. Our estimate nearly approaches such a value with a tenth of the upper estimate's mean, i.e. $11.2 - 37.5 \times 10^6$ kg (tenth of [(8)], cf. end of Sections 6.4.2). This value still ranges at the lowermost extreme within this upper limit's uncertainty ([(10)] and [(11)] in Table 3). However, the amount of the daily influx of meteoritic material is a matter of debate and could be a tenth (Plane, 2012) of what is specified by other references (Love and Brownlee, 1993; von Zahn, 2005)."

**Referee Comment:** Line 873: should read "consist of, or contain, non-volatile material"

➔ **Autors reply:** corrected as suggested

**"**Inside the Arctic vortex up to 8 of 11 particles with diameters larger than 10 nm and smaller than about 1 µm were observed to consist of, or contain, non-volatile material."

**Referee Comment:** Equation 1: state how the mid-latitude N2O was chosen at the different theta levels.

➔ **Autors reply:** following sentence is added in the text right after Equation 1.

"The parameterization of $N_2O_{mid\text{-}lat.}(\Theta)$ as used here is based on further measurements with HAGAR at mid-latitudes."

**Referee Comment:** Page 10: Figures 2 – 4 are not properly discussed in the text. Some of the panels are not referred to at all.

➔ **Autors reply:** In Section 4 the reference to all shown sub-figures are completed.