We would like to thank the anonymous reviewer for reading this manuscript and offering suggestions for improvements. In the following, we respond to his/her comments.

 The input of interplanetary dust particles to the middle atmosphere has a large uncertainty (a factor of 10) (Plane, Chem. Soc. Rev., 2012, and references therein). The inter-annual variation of the resultant nanometre-sized meteoric smoke particles is also unknown in the winter Arctic vortex. Thus, an assumed value of 7.5 cm⁻³ for sulfate particles having an insoluble material is a large unknown factor. Also, the composition of such a material is not evaluated well, although there is a literature in the mesosphere (Hervig et al., JASTP, 2013). The heterogeneous nucleation rate might be depending on compositions of the material. To what extent do these uncertainties affect a real nucleation rate?

We agree with the reviewer that number densities, sizes and composition of interplanetary dust particles comprise a large uncertainty. However, observations during the RECONCILE campaign showed that around 80 % of the aerosol observed in the polar vortex contained non-volatile residuals (von Hobe et al., 2013), which supports the findings by Curtius et al. (2005) reporting a number of 67 % inside and 24 % outside the polar vortex. We chose to use a non-volatile fraction of 50 %, which corresponds to 7.5 cm⁻³ of the background aerosol having an insoluble core. This value represents the mean of the large range of values reported in the literature and compared to the observations within the RECONCILE winter is a conservative estimate. This is however not a critical parameter in our NAT and ice nucleation parameterizations. Other values could be used and would produce similar results once the parameterization is slightly re-tuned. As described in Hoyle et al. (2013), only a small fraction of the meteoritic material present can act as NAT/ice nuclei, if the observations are to be reproduced. This fraction is determined for NAT by the three parameters α_0 , γ' and P_{pre} and for ice by the two parameters α_0 and P_{pre} . We expect that the relative values of these parameters will change in the future as more information on composition and number density of the insoluble material in the polar stratosphere will become available. However, any revised set of parameters will again need to also reproduce the results shown in the current papers.

Freezing temperatures and therefore heterogeneous nucleation rates depend on composition as laboratory measurements show (e.g. Hoose and Möhler, 2012, and references therein). However, as our parameterization is constructed so as to reproduce observed NAT PSC properties throughout the vortex during several months of the 2009/2010 winter, the effects of aerosol composition on NAT nucleation are accounted for in the parameterization. Interannual changes in aerosol composition could lead to changes in the ability of the parameterization to predict NAT nucleation, however we are not aware of any observations suggesting a large variability of this kind exists in the polar stratosphere.

We have refined the following text to Sect. 2.3.1 (page 8842, line 3) describing the heterogeneous ice nucleation within ZOMM:

"Our parameterization of heterogeneous nucleation does not discriminate between different kinds of dust or other solid cores that might be immersed in the stratospheric background aerosol. Not only the composition, also number densities and sizes of interplanetary dust particles comprise a large uncertainty (Plane, 2012). However, our choice of number densities and sizes are in general agreement with studies of meteoritic material, transported from the mesosphere down into the polar vortex. With an average extraterrestrial mass influx of 20 to 100 tons per day (Cziczo et al., 2001), which compares with 160 tons per day of sulfur influx from the troposphere (or 650 tons per day of aqueous sulfuric acid) during volcanically quiescent times (SPARC, 2006), meteoritic material constitutes 3 to 15 wt% of the stratospheric aerosol. It is spread globally and funneled into the polar winter stratosphere of both hemispheres by the Brewer–Dobson circulation. For our study, we assume a number density of 7.5 cm⁻³ of meteoritic particles uniformly distributed throughout the Arctic stratosphere, which results in 50 % of the total background aerosol droplets carrying meteoritic particles. These numbers are a conservative estimate compared to Curtius et al. (2005) and similar measurements

performed within RECONCILE (von Hobe et al., 2012). Stratospheric H_2SO_4/H_2O particle concentrations range from 10 to 20 cm⁻³, and a higher fraction of nonvolatile compounds was measured by Curtius et al. (2005) inside (67%) the vortex than outside (24%), supporting the funneling effect mentioned above. The foreign nuclei within ZOMM are represented with a fixed radius of 20 nm following Hunten et al. (1980), who modeled the recondensation of ablated meteoric material into nanometer-sized smoke particles. Since only a small fraction of the foreign material are assumed to serve as heterogeneous nuclei (compare Hoyle et al, 2003), a change in number densities or radii of the material present has no effect on the conclusion of this study. A slightly re-tuned parameterization would produce similar results for different percentages of non-volatile residuals."

2. For the NAT nucleation, the authors do not mention the possibility of pseudoheterogeneous processes (e.g., Tabazadeh et al., JPC, 2002). Why is this excluded in this study? A recent laboratory study suggests that ice crystals can be coated by super-cooled ternary or binary solutions (Bogdan et al., Nature Chem., 2010). How is the impact of this on the homogeneous ice nucleation or the heterogeneous NAT nucleation on ice? SAT can also be formed from sulfuric acid solutions containing soluble meteoritic metal (Wise et al, JGR, 2003). Is such the SAT particle not a candidate for ice/NAT nuclei? In addition, the cosmic ray induced nucleation should also be discussed (Yu, ACP, 2004). Still need more discussion regarding possible nucleation pathways.

First, Tabazadeh et al. (2001) argued that homogeneous nucleation of NAD and NAT in liquid aerosols under polar stratospheric conditions might lead to denitrification, which was refuted by Knopf et al. (2002). Next, Tabazadeh et al. (2002a) discussed the possibility of surface-based nucleation of NAD and NAT from liquid ternary aerosol. They reanalyzed experimental data on homogeneous nucleation rates, which had so far been discussed against the background of volume-based nucleation. Although they argued for a surface-based production rate of NAD, which was a factor of 100 higher than the volume-based rate, even such a rate would remain too small to explain observed number densities of HNO₃ containing particles in the stratosphere (Knopf et al., 2002; Stetzer et al., 2006; Möhler et al., 2006). Pseudoheterogeneous nucleation rates for NAT remain lower than those of NAD. Moreover, direct observational evidence for NAD in the stratosphere is missing (Lowe and MacKenzie, 2008), and one of the few measurements of the H₂O:HNO₃ ratio in PSC provides ample of evidence for 3:1 (NAT) and 5.5:1 to 6.5:1 (STS), but no evidence for 2:1 (NAD). The same applies for SAT. There is no observational evidence for the existence of SAT particles (Peter and Grooß, 2012). Furthermore, although it is likely that SAT heterogeneously nucleates on other crystalline solids in the stratosphere, there are no studies of nucleation rates (Lowe and MacKenzie, 2008). For these reasons, we excluded pseudoheterogeneous processes, NAD as well as SAT from our modeling study.

A coating of ice crystals by supercooled $HNO_3/H_2O/H_2SO_4$ solutions could indeed change the nucleation rate of NAT on ice (Biermann et al., 1998). So far, only NAT nucleation by vapor deposition onto ice surfaces is accounted for in ZOMM. A heterogeneous nucleation rate of NAT on ice in the immersion mode would be much smaller (e.g. Koop et al., 1995; 1997) and could hardly explain synoptic-scale areas of Mix2-enh clouds downstream of ice clouds. More freezing experiments with ternary solutions might be needed to fully understand NAT nucleation on ice. Considering homogeneous ice nucleation, we do not see how the occurrence of a residual solution coating on ice could affect the particle nucleation (because the ice is already there).

Particle nucleation induced by energetic particles (electrons, protons and heavier ions) has been discussed for cosmic rays (CRs) by Yu (2004) and for solar energetic particles (SEPs) by Yu (2004) and more recently by Mironova et al. (2008; 2012). Yu (2004) analyzed the effect of SEPs on PSCs during the Arctic winter 2000 and Mironova et al. (2012) focused on a solar proton event in 2005. Solar proton events may enhance the flux of energetic ions by many orders of magnitude at the top of the atmosphere (Mewaldt et al., 2013) and still more than one order of magnitude in the lower stratosphere (Yu, 2004). However, Yu himself states that "Further

studies are needed to either confirm or reject the cosmic ray induced freezing hypothesis" and draws a couple of years later as a co-author of English et al. (2011) the conclusion that WACCAM "contains the sulfate microphysical processes needed for simulations in the UTLS, and that the properties of particles with sizes relevant to climate, cloud physics and heterogeneous chemistry are not sensitive to the details of the nucleation scheme or to the presence or absence of ion nucleation." Also the studies by Mironova et al. leave room for uncertainty since her conclusions are based on "all latitude data", which is a complex mixture of outof-vortex and in-vortex aerosols and PSCs. Additionally, the extended solar minimum from January 2007 until the middle of 2010 led to very low fluxes of SEPs and no solar proton events during the RECONCILE winter (Mewaldt et al., 2013). Conversely, the cosmic ray (CR) intensity is higher during solar minima, because the solar wind is then weaker and does not deflect the CRs. However, in contrast to the order-of-magnitudes differences in fluxes of energetic particles in solar proton events, the ratio in CR intensity between solar max and solar min is at most a factor 2 at the top of the atmosphere (Mewaldt et al., 2013) and in the lower stratosphere (Calisto et al., 2011). While a general effect of CRs on NAT or ice nucleation cannot be excluded and needs to be examined in future work, it seems very unlikely that a factor-of-2 increase in CR intensity during the RECONCILE winter could play any discernible role.

In summary, there is little evidence to support nucleation induced by energetic solar or galactic ions playing as significant a role in PSC formation as the heterogeneous nuclei observed in Arctic stratospheric aerosol particles (Curtius et al., 2005; von Hobe et al., 2013) likely do.

A more detailed discussion regarding possible NAT nucleation pathways can be found in the companion paper. In order to account for the reviewer's comments and focusing on possible ice nucleation pathways, we extended the discussion within the Introduction (page 8834, line 21) as follows:

"Even though 2009/2010 was an Arctic winter with unusually low minimum temperatures, we show here that these temperatures are, in themselves, insufficient to explain the CALIOP ice observations in terms of homogeneous nucleation. Rather, ice nucleates homogeneously only when $T < T_{\text{frost}} - 3$ K (Koop et al., 2000), which according to meteorological temperature data was clearly not reached on synoptic scales. The volume based nucleation rate coefficient suggested by Koop et al. (2000) aligns with classical nucleation theory and various laboratory studies analyzing ice nucleation within binary and ternary solutions (e.g. Middlebrook et al., 1993; Koop et al., 1998; Chang et al., 1999), whereas the importance of surface-based ice nucleation proposed by Tabazadeh et al. (2002b) has not been confirmed.

Since heterogeneous nucleation of NAT is necessary to explain the CALIOP observations in December (when temperatures stayed more than 5 K above $T_{\rm frost}$ – 3 K as shown in Fig. 1 by Hoyle et al., 2013), this suggests that a similar pathway might exist also for ice formation. For the troposphere, different laboratory as well as theoretical studies show that this process is of importance for ice cloud formation (e.g. Zuberi et al., 2002; DeMott et al., 2003; Kärcher and Lohmann, 2003; Cziczo et al., 2013 and references therein). However, little attention has been paid to the implications of heterogeneous ice nucleation for PSC formation, although Bogdan et al. (2003) have shown that fumed silica, possibly representative for meteoritic smoke particles, is suitable to induce heterogeneous freezing of ice under stratospheric conditions. In addition to heterogeneous nucleation of ice on foreign nuclei, the possibility of heterogeneous nucleation on preexisting NAT particles will be investigated. The nucleation of ice on sulfuric acid tetrahydrate (SAT) has also been discussed in the past. However, we do not further investigate this potential ice formation pathway due to unknown formation routes and lack of observational evidence for the existence of SAT (e.g. Lowe and MacKenzie, 2008; Peter and Grooß, 2012). Also, the early onset of NAT formation in December 2009, when the presence of SAT was very unlikely, demands a NAT formation mechanism which can hardly be SAT-induced. Finally, ice or NAT nucleation caused by galactic cosmic or solar energetic particles penetrating STS droplets as proposed by Yu (2004) is not considered in this study. An extended solar minimum with very low fluxes of solar energetic particles ranging from January 2007 until the middle of 2010 (Mewaldt et al., 2013) turns this option into an unlikely possibility to explain the observations. Galactic cosmic rays remain a possibility that we cannot exclude, but their interannual variability is weak and their microphysical mechanism remains more speculative than the reference

to heterogeneous nucleation on the observed undissolved nuclei in the stratospheric background aerosol (for more details see also the Interactive Discussion on this paper)."

Minor comments:

1 Introduction

page 8833, line 15, Is this reference (Solomon, Nature, 2004) appropriate for this explanation?

Solomon (2004) gives a rather detailed explanation for synoptic-scale temperatures, which are on average warmer in the northern than in the southern winter polar hemisphere: "Ultracold temperatures are far more widespread and persistent in the Antarctic winter and spring atmosphere than in the Arctic, where the flow of air over the Himalayas and Rocky Mountains and land—sea temperature contrasts can generate very large 'atmospheric waves'. On the ground, we experience many of these waves as the passage of storms, and some travel upwards to the stratosphere, ultimately mixing warmer mid-latitude air with cold polar air. So the varied topography of the Northern Hemisphere gives it a greater number of atmospheric waves and a warmer polar stratosphere in the winter and spring on average than in the south."

We added the original citation given by Solomon (2004) as a reference in our manuscript, namely WMO (1998), and clarified the text (page 8833, line 13) as follows:

"...owing to the larger land-ocean contrasts in the Northern Hemisphere generating atmospheric waves, which weaken the Arctic polar vortex, lead to enhanced mixing of warmer air masses from lower latitudes into the polar vortex and increase the synoptic-scale temperatures (WMO, 1998; Solomon, 2004)."

page 8833, line 22, Provide a reference for the CALIPSO observation.

We added Winker et al. (2009) as a reference.

page 8834, line 24, What is a reference for the homogeneous nucleation of ice at T below T(frost) - 3K?

We added Koop et al. (2000) as a reference.

2.2 Trajectory calculations

page 8837, line 26, How do you select the PSC free area?, since MLS does not capture PSC.

We selected the PSC free areas with the help of nearly coincident CALIOP PSC observations. Spatial and temporal differences between MLS and CALIOP are less than 10 km and 30 s after a repositioning of the Aura satellite in April 2008 (Lambert et al., 2012). We added this information to the manuscript.

page 8838, line 9, In addition to the measurement uncertainties of MLS, spatial resolutions of MLS also contribute to the modelling uncertainty. Differences between MLS and CALIOP resolutions (vertical and horizontal) should be stated.

Vertical along-track resolutions are 3.1 km to 3.5 km for H_2O , and 3.5 km to 5.5 km for HNO_3 . The horizontal resolution is 180 km to 290 km for H_2O and 400 km to 550 km for HNO_3 . Because we are using vortex-averaged MLS data (see above), we added as additional information for the reader only the vertical resolution at page 8837, line 27. Please see also the first comment of the second review.

2.3.2. Heterogeneous NAT nucleation

page 8843, Please add definitions for gamma and gamma(prime).

We changed the text within the corresponding section as follows:

Our current understanding of PSC formation includes two mechanisms to nucleate NAT. First, the nucleation scheme of NAT particles forming on solid inclusions such as meteoritic dust is described and discussed in detail in the companion paper by Hoyle et al. (2013). Second, the original approach, the formation of NAT on preexisting ice particles, which follows Luo et al. (2003). The parameterization for the nucleation rate for NAT on ice is defined as follows:

$$J_{\rm NAT}(T) = 6.24 \times 10^{24} \rm cm^{-2} s^{-1} \times (T/K) \times exp \left[\frac{273.15^3}{T^3} \frac{\gamma}{(\ln S_{\rm NAT}(T))^2} - \frac{2000 \rm K}{T} \right].$$

The parameter γ was constrained by Luo et al. (2003) to be

$$\gamma = \frac{16 \pi m^2 \sigma^2 f}{3 \rho^2 k^3 273.15^3} = 328 \text{ K}^3.$$

The molecular mass of NAT is defined as *m*, while ρ is the density of NAT and *k* the Boltzmann constant. Unknowns are the surface tension α and *f*, which describes the lowering of the Gibbs energy barrier due to the presence of the ice surface. We discuss the importance of changing γ in Sect. 3. The newly developed NAT nucleation parameterization on foreign nuclei differs from Luo et al. (2003) such that γ longer includes the compatibility factor *f* and therefore the parameterization accounts for active sites of different quality. For this reason, Hoyle et al. (2013) defined γ' , used in our simulations with a value of either 650 K³ or 700 K³, depending on whether or not small-scale temperature fluctuations are accounted for.

2.4. Small-scale temperature fluctuations

Accuracy of temperature is very important. How well does the estimated temperature agree to high resolution temperatures measured by, for example, GPS radio occultations?

We compared ERA-Interim reanalysis as well as the higher resolution ECMWF operational analysis temperatures to unassimilated temperatures measured from radiosondes, which have been launched from Ny-Ålesund and Sodankylä during the RECONCILE campaign. This comparison reveals the best agreement between measured and modeled temperatures for the ERA-Interim data set. Furthermore, the comparison shows that small-scale fluctuations are not accounted for in either of the data sets and provides an estimate of vertical velocities. Fluctuation amplitudes agree with estimated temperature fluctuations superimposed onto the synoptic-scale trajectories. Only wavelengths < 400 km were considered (page 8844, line 25). This number was wrongly specified as 100 km and corrected to 400 km. Even though the grid point distance of the underlying meteorological data is 100 km, about four grid points are required to resolve a sinusoidal wave pattern.

COSMIC GPS temperatures are already assimilated in ERA-Interim (Dee et al., 2011), and therefore do not provide a totally independent data set, which could be used for comparison.

3. Results and discussion

page 8849, line 15, Why not show the boxes in a figure (as a supporting material)?

We included the boxes into Fig. 3. By doing this, we realized a typing error in the text, namely that we divided the domain defined by Mix2-enh, ice and wave ice only into n = 9 instead of 12 boxes. We corrected this on page 8849, line 16 and 18. The calculations are correct.



Fig. 3: Simulated model results for an exemplary CALIPSO orbit. Results are shown within the 2-D scatter plot of aerosol depolarization ratio ($\delta_{aerosol}$) versus inverse R_{532} ($1/R_{532}$). (a) Unperturbed model results. (b) Model results with applied CALIOP uncertainties (σ). Uncertainties in parallel and perpendicular backscatter are calculated using Eq. (6), propagated into $\delta_{aerosol}$ and $1/R_{532}$ and shown as red error bars. Yellow boxes denote a division of the particle classes Mix2-enh, ice and wave ice to quantify the performance of different model runs as discussed in Sect. 3.

page 8851, line 18, This is refer to the sedimentation of PSC particles fallen down to the lower layers. But also need some discussion about the possibility for NAT/ice particles that are falling from the above layers.

To clarify this, we added the following sentence at the end of the paragraph in line 20:

"NAT number densities might be reduced or enhanced by accounting for a vertical redistribution of large enough PSC particles, by falling out, falling through or accumulating in the corresponding cloud layer."

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