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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical analysis of refractory stratospheric aerosol particles collected within the arctic vortex and inside polar stratospheric clouds" *by* Martin Ebert et al.

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A general statement was that there are too many subjective terms (e.g. "very low") and that a careful editing is necessary. We have performed this editing process, in which we have tried to eliminate all subjective terms and to specify our statements.

Beside this general point, four detailed points has to be answered:

Point 1 (Introduction Page 2, Paragraph starting "One source" : Suggest that the work of Murphy et al., Science, 1998 and Murphy et al., Carbonaceous material in aerosol particles in the lower stratosphere and tropopause region, JGR, 2007 be referenced here. Murphy quantified many of the sources and their effect on the UTLS that are

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Discussion paper

described here and that work predate that referenced here.).

We agree - We placed the references of Murphy et al. (1998 & 2007) in the opening part of the introduction.

"One major source region of stratospheric refractory particles is assumed to be the interplanetary space [Murphy et al., 1998 & 2007; Plane, 2012] from where cosmic dust is incorporated into the Earth's atmosphere."

Point 2 (Section 4.1.3 : It seems odd that the authors discuss large silicate particles as being non-chondritic in composition and use this to dismiss a meteoritic origin. High temperature processing is known to lead to loss of more volatile materials (e.g. Mg over Si) and that residual spherules are the remains of heated but not fully ablated material (the authors should see e.g. F. J. M. Reitmeijer, G. J. Flynn, Meteorit. Planet. Sci. 35, A136 (2000)). It is therefore unclear why this assumption has been made; it should be better quantified or reconsidered. The authors should go back and read the literature in this area.):

Reviewer 2 indicates the fact that meteoric composition within individual grains is known to change during high temperature processing and references the work of Reitmeijer (2000). The authors are aware of this fact and also know the overview paper of Reitmeijer. Because of the fact that such volatility studies has shown that if the mass loss is not bigger than 80% , the Mg/Si ratio is not changing so much (Floss et al., 1996), we had used this ratio as marker for chondritic material. But indeed, this studies also show that if mass loss is bigger than 80% Mg becomes strongly depleted within the residues and in this way also the Mg/Si ratio is no good marker anymore. Therefore we have completely rewritten this section.

New text in chapter 4.1.3 "In stratospheric samples silicates are often described as main refractory component. As source for these silicates most often chondritic material from cosmic dust is discussed. The original chondritic material is characterized by specific elemental ratios, e.g. a high Mg/Si ratio (∼1, Rietmeijer (1998)). Similar values

(∼ 0.85) were determined for the small (particle diameter < 500 nm) silicate spheres found during ESSenCe and RECONCILE. The larger silicate spheres (particle diameter > 500 nm), which were only found in the RECONCILE samples, show lower Mg/Si ratios (∼ 0.4). It is known that high temperature processing leads to a loss of more volatile materials. Volatility studies have shown that within the residues up to a remaining fraction of 20% Mg and Si behaves very similar, but for even higher ablation rates Mg becomes depleted in contrast to Si (Floss et al., 1996). In this way the residual spheres as the remains of heated but not fully ablated material can show completely different elemental ratios. In this way Riemtmeijer (2000) states: "We may consider the possibility that nonchondritic element abundances are a norm rather than an exception." Twenty percent of the large silicate particles during RECONCILE are not spheres but show irregular morphologies (Figure 3f). Even when these fact does not allow an unambiguous assignment to a high temperature history, these particles can originate from cosmic dust. A different source for such particles is discussed by Zolensky et al. (1989). They suggested that such silicates with nonchondritic composition (which were the majority in the size range 1 – 5 μm in their study) may originate from aerospace activities. They listed as sources solid rocket fuel exhaust, solid rocket motor ablation, thermal reflective paint from the outer hull of spacecraft, and ablating hardware from satellites and discarded rocket sections in low-Earth orbit."

Point 3 (Section 4.1.8): Section 4.1.8 Aluminium oxide spheres: This section would be stronger if enhanced with some of the more common references in this field. The authors references the modeling study of Jackman et al., 1998 and the size distribution of . Suggest looking at (and referencing) the more comprehensive work of Danilin, M. Y., et al., Global stratospheric effects of the alumina emissions by solid fueled rocket motors, J. Geophys. Res., 2001 and the compositional work of Cziczo et al. Composition of individual particles in the wakes of an Athena II rocket and the space shuttle, GRL, 2002. Again,there is considerable literature in this area – indeed, NASA ran an entire program on the effects of these emissions on the upper atmosphere that is not referenced – that needs to be reviewed. Also, were there rocket launches that might have led to the observations? Can the authors support their assumptions with any ancillary data e.g. back trajectories?

We have added a variety of citations with more comprehensive work. Also we now cite the CIAP program and we refer to rocket launches during the RECONCILE campaign.

New text in chapter 4.1.8 “Solid rocket motors respectively the impact of launch vehicle activity are known to be a source for aluminum rich particles in the stratosphere (e.g. Cziczo et al., 2002). The impact of these particles on the heterogeneous chemistry of stratospheric ozone is studied since the early 70‘s as part of the Climatic Impact Assessment Program (CIAP) (Hoshizaki, 1975). Detailed discussion can be found e.g. by Danilin et al. (2001), Jackman et al. (1998) and Denison et al. (1994). Cofer et al. (1991) measured a bimodal size distribution of AOS in the Space Shuttle plume with peaks at 2 μm and < 0.3 μm. Small AOS spheres (diameter around 100 nm) were observed during RECONCILE and ESSenCe (see Figure 7a). While no externally mixed supermicron AOS particles were detected during both campaigns, supermicron AOS were part of the complex agglomerates within the mixed metal/ metal oxide group, observed during RECONCILE and described in chapter 4.1.5. In Kiruna rockets are launched within the framework of the ESRANGE program. A local plume from this activity cannot be expected as the last rocket launches from Kiruna prior to RECONCILE were conducted at 22th and 29th of November 2009 and in this way around 7 weeks before the sampling flights. But there is a constant entry of particles from space launch activities in stratosphere and mesosphere. For example, during the time period of RECONCILE at least 9 space launches took place, including Proton rockets, Soyuz-U, Delta rockets, and the Space Shuttle Endeavour (see www.spacelaunchreport.com/log2010.html).”

Point 4 Regarding nucleation in PSCs : the unsupported statement that ends the paper “Thus,the observed refractory particles seem to provide a surface for heterogeneous condensation during PSC events.” is another example where there exists considerable literature (starting with the work of Biermann et al., GRL, 1996) that would make this case. Absence of citation of previous work should not occur.

For a long time there is the discussion in which way heterogeneous nucleation plays a role in PSC formation. It was not our goal in any way to present this as a new idea. Therefore, we have rephrased this point and we have added some citations.

New text in Conclusion: “Refractory particles can provide a surface for heterogeneous condensation during PSC events. The occurrence and importance of this possible PSC nucleation pathway is controversial discussed and tested in laboratory experiments in the last years (e.g. Biermann et al., 1996; Engel et al., 2013). While the data base is still sparse, we have observed the general tendency that the number of large refractory particles, which were encountered during the special conditions of RECONCILE, decrease during PSC events. We explain this experimental observation by the activation of the large refractory particles into PSC elements under favorable conditions. In this situation sampling of these particles by the fast-flying Geophysica aircraft is prevented due to particle and subsequent inertia growth.

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