Atmos. Chem. Phys. Discuss., 13, C3746–C3749, 2013
www.atmos-chem-phys-discuss.net/13/C3746/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous formation of polar stratospheric clouds – Part 2: Nucleation of ice on synoptic scales" *by* I. Engel et al.

Anonymous Referee #1

Received and published: 15 June 2013

This is a companion paper by Hoyle et al. (ACPD, 2013) and focuses on ice nucleation in the Arctic lower stratosphere. A motivation of this paper(s) comes from an observational evidence from CALIOP that NAT clouds were formed through a non-ice nucleation mechanism (Pitts et al., ACP, 2011). A newly developed ZOMM in which incorporates heterogeneous nucleation of NAT and ice could fairly reproduce characteristics of the CALIOP measurements. As the authors stated, a three dimensional model simulation considering mixing of airmass and sedimentation of PSCs is highly expected in a further study. But, this study is an important step for future works (whether the mechanism presented here can be applied for other area such as the Antarctic or

the mesosphere). I therefore recommend that this manuscript is published. Following questions and comments should be addressed before the publication.

1. 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 depend on compositions of the material. To what extent do these uncertainties affect a real nucleation rate?

2. For the NAT nucleation, the authors do not mention the possibility of pseudo-heterogeneous 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.

Minor comments:

1. Introduction

page 8833

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

line 22, Provide a reference for the CALIPSO observation.

page 8834

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

2.2 Trajectory calculations

page 8837

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

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.

2.3.2. Heterogeneous NAT nucleation

page 8843

Please add definitions for gamma and gamma(prime).

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?

3. Results and discussion

page 8849

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

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 8831, 2013.