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

Anonymous Referee #2

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This paper investigates the significance of heterogeneous nucleation on observations of solid-phase Type I NAT/PSC in the northern hemisphere during winter 2009/10, and is a companion paper to Hoyle et al. (Part I - AMT). Nucleation pathways for PSC, both Type I and II, have long been studied, and the importance of heterogeneous nucleation makes relevant the types of active ice nuclei available in the upper atmosphere. A recent paper by Czizco et al. in Science (2013) shows the potential underrepresented importance of this process in the troposphere. This paper makes a similar case in the lower stratosphere. The paper is thus very relevant to ACP. Figures are very clear and suitable for publication, and the manuscript is well-written and fairly straight-forward to

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read/understand.

This is the first time this reviewer has seen this manuscript.

My summary recommendation to the editor is to accept this paper under the condition of minor revision.

My major concerns are:

- Please tone down hyperbole ('unprecedented' in the abstract) and be more circumspect in your conclusions so as to be more consistent and considerate of your (relatively limited) experimental design. There are large uncertainties in your methods, despite your presenting a relatively compelling story and narrative. In particular, your lack of constraint for H₂O and HNO₃ mixing ratios is very concerning. MLS is coarse, and your interpolation schemes even coarser. My estimates say that the uncertainties in your frost point estimates likely exceed 2 K, which is enough to weaken many of your claims (you should be more forthright about these uncertainties). Additionally, given the recent Nature paper by Anderson et al. (2012), and the potential enhancement of NH water vapor from increased overshooting midlatitude convection, the uncertainties in H₂O are potentially very large. Back trajectories, particularly in the lower stratosphere, are further limited by uncertainty. I'm not generally questioning the veracity or fidelity of your claims and conclusions, but I don't think should be accompanied by such bold claims as 'unprecedented'.

- I'm having difficulty reconciling the claim that "This is in contradiction to our previous laboratory-based understanding of NAT formation, which (1) excluded the possibility homogeneous NAT formation" with the model of Tabazadeh et al. (1994 - GRL) that seemingly opens the possibility for this path.

- It would be useful for the reader to justify your claim that ice nucleates homogeneously only when $T \leq T_{\text{frost}} - 3\text{K}$, as I believe that you are attributing this to the rate of necessary vapor supersaturation. Its best clarified for the reader.

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- Your reconciliation of meteoric materials as the source of heterogeneous ice nuclei (btw, please be clear and up front that you are discussing contact nucleation almost exclusively...it comes out later in Sec. 2.3.1, but was unclear my first go through the text) is fairly unconvincing. Its mostly hand waving to suggest that your model parameterizations seem most in line with observations and then use words like "likely" in the abstract. Can you change that to "plausibly" to be more neutral (again, that word circumspect)? In fact, in the conclusions, the narrative is more circumspect on this point. Food for thought, I'm frankly very interested in the impact that pyrocumulonimbus residues have on background stratospheric aerosol particle fields (non-volcanic). Surely, biomass burning materials aren't as efficient as active ice nuclei as other materials, but you guys make some assumptions about the distribution of the background composition that doesn't necessarily consider active perturbations like PyroCB (see Fromm et al., 2010 BAMS).

- I'd like to see your conclusions broadened. Where are you going with this work? What is the significance of heterogeneous nucleation being potentially dominant relative to homogeneous? How does this effect potential and future parameterizations in your model? Ultimately, what is the point? This is very good paper, and you guys just sorta let it die when you should seemingly have so much more to say about impact and future directions.

Good paper. Good luck!

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

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