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Comment on acp-2022-598

Anonymous Referee #2

Referee comment on "The importance of acid-processed meteoric smoke relative to meteoric fragments for crystal nucleation in polar stratospheric clouds" by Alexander D. James et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-598-RC2, 2022

The paper by Alexander James and colleagues addresses an important and highly up to date topic. The presence, the amount, and the importance of meteoric material in the stratosphere is uncertain but discussed in recent studies to explain observations of polar stratospheric clouds (PSCs). Any piece of information from measurements and laboratory studies is welcome to solve this puzzle. The study by James et al. delivers such new and interesting findings supporting ideas about heterogeneous cloud formation on meteoric material in the polar winter stratosphere. However, the study is in places difficult to understand and not always stringent. I would like to point out one general difficulty that I had with the paper before I list a couple of specific comments.

General comment:

You come to the conclusion that meteoric fragments can nucleate nitric acid hydrate crystals, however, there is unlikely to be sufficient input flux to explain observed crystal numbers in PSC. You further propose that acid processed meteoric smoke can also nucleate NAT, despite its small size. In contrast to meteoric fragments, observed crystal numbers in PSCs can be explained by the input flux of meteoric smoke. The first question I have is, why do you extend your previous setup from your study in 2018 by adding H_2SO_4 only for the experiments with meteoric fragments? Why don't you do this not also for meteoric smoke which seems to be much more relevant for PSC formation? The second request I do have: I would like to see equal figures for both types of meteoric material. I would like to compare meteoric fragments and meteoric smoke under the same conditions by looking at the same figures. Let's take Fig. 3 and Fig. 9: I would like to see the same figure with the same axis labels with the same axis numbers with the same caption and so on. The only difference should be the data. And here comes again the question: Why is it not possible to measure MSP analogues in a ternary $H_2SO_4/HNO_3/H_2O$ solution? This would increase the value of this study considerably.

Specific comments:

Introduction: Please revise your general PSC introduction completely. It contains the most important PSC facts but the details are not thoroughly written down. The selection of references could be better, too.

Page 2, Line 36: I would add more citations, e. g. Manney et al. (2020), Wohltmann et al. (2020), Dameris et al. (2021). Lawrence et al. (2020) is particularly about the vortex strength.

Page 2, Line 42: In my view, Wegner et al. (2012) is not appropriate here. Please search for a better reference related to denitrification. I would suggest Crutzen and Arnold (1986).

Page 2, Line 43 ff: The sentence "In many clouds NAT is thought to nucleate on ice crystals, but it has been shown that crystalline nitric acid particles can sometimes form in conditions where ice is not thermodynamically stable (Mann et al., 2005; Tritscher et al., 2021)" contains a valuation which is probably not on purpose. It sounds like the majority of NAT particles is formed through the nucleation on ice crystals and only "some" NAT particles form via different pathways. I would express this in a more neutral way because this weighting might be wrong from today's perspective.

Carslaw et al. (1999) shows results from a model study stating that mountain-induced mesoscale temperature perturbations may be an important source of nitric acid hydrate particles in the Arctic. He does not claim that "all" clouds contain water ice. In 1999, he was still missing the vortex wide picture by satellite observations. Also the model resolution was quite coarse at that time (5 x 2 degree). Therefore I do not support your statement "though in some winters effectively all cloud could contain water ice" – in general and also not with this citation.

Page 2, Line 50 ff: "Global models do not yet include a parameterisation of this activity based on laboratory measurements of reasonable heterogeneous nucleator surfaces, but rather have been tuned to observed NAT particle concentrations (Grooß et al., 2014; Voigt et al., 2005)." You cite Grooß et al. (2014), however, this paper contains a NAT parameterization based on the active site theory. Also Zhu et al. (2015, 2017a,b) describes the very detailed PSC scheme of WACCM. On the other hand, you cite Steiner et al. (2021) which is a global model. However, you cite it together with box models. Please have a look at Table 7 from Tritscher et al. (2021) to find out more about PSC parameterizations in models and revise this paragraph.

Page 2, Line 55: Figure X1 ☐ Figure 1

Figure 1: Your y-Axis (Altitude) goes down to 0, even below. The H_2O concentrations in the troposphere are higher than 5 ppm! Please draw the lines for H_2O, HNO_3 and H_2SO_4 concentrations more carefully/exact.

Within the introduction, I would like to see a clear definition of meteoric smoke and meteoric fragments. Please explain the differences between smoke and fragments such that a reader who is not as familiar with the topic and nomenclature as you are can easily understand it. This is important to get the main massage of your paper! It is also important to use this terminology through the whole paper. If you introduce an abbreviation, like MSP, please use this abbreviation everywhere and do not switch back to the write out version, this is confusing.

Page 8, Line 153: Could you please explain in more detail where the 0.5 wt% H_2SO_4 is coming from?! If I look at the corresponding plot (Fig. 12 in Carslaw et al., 1997), I assume a value higher than 0.5 wt%. Looking at Tab. 1 in Biermann et al. (1996), based on Carslaw et al. (1994), it would be at 190.0 K a value of 41.2 wt% for HNO_3 and 3.9 wt% for H_2SO_4. Even below the frost point, the concentration of H_2SO_4 is still at 2.5 wt%.

Page 10, Figure 3, Caption: Please make one or two complete sentences at the beginning. "are data from (James et al., 2018)" change to James et al. (2018)

Page 12, Figure 4: Please describe in the caption of the figure always the abbreviations used in the figure, here n_s and S_{NAT}. It is more convenient for the reader to find it in the caption instead of searching the main text body.

Page 13, Line 230: Where does the 6×10^{-6} cm^{-3} comes from? Reference?

Page 17, Line 318: 3:1 H_2O:HNO_2 change to HNO_3

Page 22, Line 413: Change 0.236 mJ m^{-2} in 0.236 J m^{-2}

Page 25, Line 450f (Caption of Fig. 10): Change ppmv in ppbv HNO_3

My impression is that you need to check carefully when you use italic fonts. Sometimes, the unit is in italic fonts, sometimes the variable and sometimes none of both.

Please distinguish more carefully surface area and surface area density (e. g. y-axis Fig. 6 is surface area density).

Please use abbreviations contentiously throughout the manuscript and don’t switch forth and back with using them or not.

Additional references that were not listed in the paper itself:

Biermann, U. M., Presper, T., Koop, T., Mößinger, J., Crutzen, P. J., & Peter, T. (1996). The unsuitability of meteoritic and other nuclei for polar stratospheric cloud freezing. Geophysical Research Letters, 23(13), 1693–1696. https://doi.org/10.1029/96GL01577

Carslaw, K. S., Luo, B. P., Clegg, S. L., Peter, T., Brimblecombe, P., & Crutzen, P. J. (1994). Stratospheric aerosol growth and HNO3 gas phase depletion from coupled HNO3 and water uptake by liquid particles. Geophysical Research Letters, 21(23), 2479–2482. https://doi.org/10.1029/94GL02799

Crutzen, P. J., & Arnold, F. (1986). Nitric acid cloud formation in the cold Antarctic stratosphere: A major cause for the springtime “ozone hole”. Nature, 324, 651–655. https://doi.org/10.1038/324651a0

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Manney, G. L., Livesey, N. J., Santee, M. L., Froidevaux, L., Lambert, A., Lawrence, Z., et al. (2020). Record-low Arctic stratospheric ozone in 2020: MLS observations of chemical processes and comparisons with previous extreme winters. Geophysical Research Letters, 47(16). https://doi.org/10.1029/2020GL089063

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