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Referee comment

Anonymous Referee #1

Referee comment on "Large hemispheric difference in nucleation mode aerosol concentrations in the lowermost stratosphere at mid- and high latitudes" by Christina J. Williamson et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-22-RC1, 2021

This manuscript investigates the number concentrations and origin of aerosol particles, especially in the sub-12 nm size range, in the lowermost stratosphere at mid and high latitudes. The investigation is based on air craft measurements augmented by theoretical arguments and box model simulations. The topic is important and original enough to warrant publication. The paper is well organized and clearly written. I have a few, mostly minor, issues that should be addressed before I would recommend acceptance for publication. My detailed comments in this regard are summarized below.

Scientific comments

The size range 3-12 nm is called sometimes ultrafine particles, sometimes nucleation mode particles in the paper. I would strongly recommend avoiding the term ultrafine particles in this context, as the vast majority of air pollution scientists use this term for the whole <100 nm particle population. The authors have also applied the size ranges 3-12 nm and 12-60 nm for the nucleation and Aitken mode, respectively. In most studies conducted in the lower troposphere, the border between the nucleation and Aitken mode has been assumed to be somewhere in the range 20-30 nm, while the Aitken mode has been assumed to extend up to 90-100 nm. The lower size ranges for these modes applied here are acceptable because the whole particle population seems to be shifted to smaller sizes, possibly due to lower concentration levels of aerosol precursors, compared with the lower troposphere. However, due to the somewhat unusual definitions of nucleation and Aitken mode size ranges, I suggest that the author add a couple of lines into the text to explain why they used such definitions for the nucleation and Aitken modes.

Somewhat related to the previous comment, it is not a good practice to talk about small particles (e.g. title of section 3) or larger particles (lines 438 and 481) without specifying what is exact meant by small or larger here.

In several places of the text, the authors talk about correlation and even its character (significant, slight). Technically, correlation is an exact statistical quantity, which should be used just based on visual the appearance on how two variables seem to be connected with each other. I recommend using some other term than correlation in the text or, alternatively, to calculate the actual correlation coefficient and its level of significance.

Line 361: The lifetime of nucleation mode particles depends both on the mean size of these particles and on the properties of the pre-existing particle population at sizes larger than the nucleation mode. Considering that both of these quantities are probably quite variable in the LMS, and especially quite different between NH and SH (as discussed in the supplementary material and illustrated in Figure S3), the authors should better justify the use of a single lifetime of 2 days for nucleation mode particles in their calculations.

I am basically fine with the way this paper discusses and speculates about the particle origin in the study regions, including the mechanism of new particle formation. The only thing that could be improved the analysis of probability of (ion-induced) water-sulfuric acid nucleation is evaluated here based on the model framework developed by Kazil, Lovejoy and co-workers 10-20 years ago. Since then, detailed laboratory data on the same nucleation mechanisms has been obtained e.g. in CLOUD experiments, and these data have also been included in nucleation parameterizations. I am not saying that the authors should redo their calculations, but they could shortly discuss whether their conclusions are also consistent with this most up-to-date information on atmospheric water-sulfuric acid nucleation at cold temperatures.

Minor, mostly technical issues

Line 83: The authors could add the study by Sipila et al. (2016, Nature 537, p. 532), because it is the very first study in which iodine compound have been measured in molecular clusters associated with new particle formation in a coastal atmosphere.

Lines 193-194: Stating that SO2 concentration measurements are sensitive to <100 pptv does not really tell anything useful to the reader. What is the actual detection limit of the instrument under the operating conditions of this study, and reliable are the SO2 of just a few pptv reported in many of the figures?

Line 243: It seems that something is missing from this text: …it is conceivable the low…

The same text is repeated on lines 319-323 and lines 325-329, except that they refer to a different figure.