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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Microphysical properties of synoptic scale polar stratospheric clouds: in situ measurements of unexpectedly large HNO_3 containing particles in the Arctic vortex" *by* S. Molleker et al.

Anonymous Referee #1

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General Comments

This paper presents in-situ measurements and supporting analyses of large polar stratospheric cloud (PSC) particles collected in the Arctic in January 2010 and December 2011 by instruments onboard the M-55 Geophysica aircraft. Inferred particle sizes and number densities are larger than those of the well-known "NAT rock" observations of Fahey et al. (2001) and cannot be explained by conventional PSC microphysical simulations assuming spherical particles with a nitric acid trihydrate (NAT) composition.

The authors offer two hypotheses that may explain their observations: non-spherical particle shapes, and/or a non-NAT particle composition, e.g. NAT-coated ice. I think the paper will be of considerable interest to the PSC and stratospheric chemistry community, but I also feel the paper is not well organized and jumps around too much between flights, instrument descriptions, and data interpretations and intercomparisons. It was (and still is) very difficult for me to keep track of which instruments are being used/compared on which flights, and it is not clear to me how some of the measurements presented relate to the major theme of the paper, i.e. unexpectedly large HNO3 containing particles. I recommend that the paper be restructured so that the reader – in particular the non-specialist– can follow the story more easily, and suggest the following structure:

1. Include a table listing each instrument whose data are being used, on which of the seven PSC flights the instrument was deployed, and a brief description of the data (e.g. FSSP-100: particle size distribution for diameters from 1.05 to 37? microns).

2. The paper jumps right into some of the observations and their interpretation in sections 2.1-2.4, which fall under the general category of measurement techniques. It would be better to first have a separate section on each instrument or at least each instrument type (where the FSSP-100, FSSP-300, and CDP could be combined), with more general discussion of what is being measured and some details on data handling, e.g. how sample areas and counting statistics come into play in the scattering probe data.

3. Then include most or all of the observations and data intercomparisons in the section on "observations and results" and try to introduce and discuss each figure in order rather than jump back and forth between earlier and later figures as is often the case in the paper in its current form.

Specific Comments

p.12073, line 7 - Use of the words "simultaneously by up to four . . . instruments" seems

misleading. Was the number of simultaneously measuring optical instruments actually two on some flights, three on others, four on some?

p.12073, line 26 - I suggest the wording be changed to “an alternate particle composition ”

p.12074, line 15 - I think the consensus is that liquid particles “likely” dominate chlorine activation, not “possibly.”

p.12075, lines 5-24 - Why not modify Fig. 1 to show temperature contours and M-55 flight tracks on actual PSC flight dates? Also, why are 30 hPa contours being shown (also in Fig. 3) when the M-55 flew at a much lower altitude? I also suggest showing ESSenCe M-55 flight tracks on Fig. 3c and Fig. 3d. Fig. 4 is not very useful; the only real information it conveys is that all flights took place from Kiruna. Fig. 4 could be eliminated if the flight tracks were included in Figs. 1 and 3.

p.12077, lines 4-9 – I think what the authors are trying to say is that the lower detection limit of the FSSP-100 was shifted so that the FSSP-100 and FSSP-300 number densities matched for particle diameters between 1.05 and 2 microns.

p.12077, lines 27-28 – I don’t understand this sentence. Do the authors mean that the STS and NAT curves are within 10% of each other in particle diameter for smaller sizes? p.12078, lines 27-28 and following – I don’t understand the concept of Depth of Field (DoF) rejected-to-accepted particle ratio. The authors need to explain what this means and why it is relevant and important to the data being presented in this paper.

p.12079, section 2.3 – The value of inter-arrival time analysis is also not clear. The authors need to re-write this section and state at the beginning of the subsection why this analysis is important.

p.12080, lines 17-21 – Fig. 10 needs to be explained much better. I don’t really understand what I am looking at in the figure. What is the importance of the 35% and 50% shadow thresholds mentioned in the caption of Fig. 10?

p.12082, lines 11-12 – What is the relevance of the MAL data shown in Fig. 11? And why do the top and bottom of the MAL data contours move up and down with the aircraft? I assume that this is an artifact of the data and not a real phenomenon?

p.12084, lines 11-22 – Again I question the relevance and importance of the MAL and MAS data shown in Fig 13. The lidar data does not really corroborate the finding that STS particles were present in large number densities. Also the lidar depolarization data indicates the present of non-spherical particles, probably on all flights, but cannot confirm the size or number density of large particles observed by the in situ optical instruments.

p.12087, lines 9-29 – I don’t understand how the chemical composition of non-volatile 0.5-5 micron particles collected during RECONCILE and ESSenCe flights is relevant to the primary message of the paper. It is interesting, however, that these particles were almost completely absent in all samples taken during PSC events. Are the authors suggesting that these particles are the primary nuclei of PSC particles? If so, they should make such a statement.

p.12091, lines 14-16 – If temperatures dropped below the frost point upwind due to lee waves over Greenland, parcel cooling rates should have been quite high, resulting in ice particles with high number density ($\sim$ 10 cm-3) and relatively small size ($\sim$ 1-1.5 microns). This seems to contradict the explanation here of the possible source of large non-NAT particles in low number densities. Could the CLaMS-based microphysical model be used to test the authors’ hypothesis about large ice particles being present downstream of the lee waves?

p.12102, Table 1 – The headings for columns 3 and 4 are identical. Should the heading for column 3 be “D > 20 microns”?

p.12110, Fig. 7 legend – I suggest moving the sentence about “. . .synoptic scale of PSCs is apparent between 46000 and 53000 UT” up to line 6 at the end of the sentence where panel (a) is introduced.

p.12111, Fig. 8 – I don't think this figure is very useful; it is included solely to establish that background aerosol measurements in 2010 by the modified FSSP-300 were similar to most data collected by the FSSP-300 in the Arctic in 1996. Is there some publication in the intervening years that could be cited to establish the accuracy of the 2010 data so this figure could be eliminated?

Technical Corrections

p.12074, line 25 – change wording to "...existence of the most stable hydrate..."

p.12074, line 2 – Does "freely floating" mean "individual"?

p.12077, line 10 – reword to "...CDP should have been at a diameter..."

p.12077, line 19 – change "objects" to "particles"

p.12078, line 19 – delete the word "of" after the word "below"

p.12084, line 28 – reword to "...particles larger than 15 nm."

p.12119, line 2 of Fig.16 legend – change "date" to "data"

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 12071, 2014.