

Reply to the reviews for the paper “Microphysical properties of synoptic scale polar stratospheric clouds: In situ measurements of unexpectedly large HNO₃ containing particles in the Arctic vortex”

We thank the anonymous reviewer and Rob MacKenzie for their constructive reviews and the diligent reading/commenting. Almost all of the suggested improvements have been implemented in the revised manuscript. Since there was a constructive reader’s comment posted by Peggy Achtert we decided to provide more details on the MAL lidar and the MAS scatterometer in a new Supplement. Some of the reviewers’ comments also lead to additional sections in this Supplement. In general, the ordering of the text and figures within the “Measurements Techniques” and “Observation and results” section also has been changed. In the following all points of reviews are cited and answered.

Comments of the anonymous Referee #1:

Reviewer Comment #1: *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).

Author’s Reply to Comment #1: In the revised version we provide a supplement, which was not part of the initially submitted manuscript. One reason is that Peggy Achert, who is not a reviewer, submitted an Interactive Reader’s Comment. We considered this comment as important contribution and drafted a detailed response to her arguments. After deciding to provide a supplement we also assembled the table suggested by Referee #1 and included it in the supplement material.

Reviewer Comment #2 and #3:

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.

Author’s Reply to Comments #2 and #3: We agree that the Section 2 could and should be improved concerning the ordering of the figures and text. Thus, Section 2.1 has been slightly rewritten with a better focus on its title, and the detection limits are given first. The text with multiple references to 3-panel figure of FSSP time series is now completely in the Results section with other references to the

same figure. In some parts of the text it is rather difficult to separate between measurement techniques and results. Nevertheless, some paragraphs and all figures were moved to the results section.

Reviewer #1, Specific comment #1: *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?*

Author’s Reply to Specific Comment #1: Good point; the wording is changed now to “by four different optical instruments”. In different flights different instruments were operating properly. This issue is solved with the Table S1 in the Supplement as proposed by the Reviewer in General Comment #1. In reality only three optical particle instruments were taking simultaneous measurements in addition to the SIOUX NO_y particle data. The major point is that operating one optical particle counting instrument fully automated on an aircraft with only the pilot on board always bears the risk of inherent subtle, undetected artefacts (like electronic noise, baseline drifts of amplifiers etc.). With several disjoint instruments partly based on different measurement principles this risk of unrealized artefacts is enormously reduced.

Reviewer #1, Specific comment #2: *p.12073, line 26 - I suggest the wording be changed to “an alternate particle composition”*

Author’s Reply to Specific Comment #2: Indeed, the suggested phrasing is better, and we corrected.

Reviewer #1, Specific comment #3: *p.12074, line 15 - I think the consensus is that liquid particles “likely” dominate chlorine activation, not “possibly.”*

Author’s Reply to Specific Comment #3: We agree and changed the sentence as proposed.

Reviewer #1, Specific comment #4: *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.*

Author’s Reply to Specific Comment #4: We discussed this among the co-authors. In principle the Fig. 4 could be omitted, by indicating the flight tracks or the measurement region in the Fig.1 and Fig. 3. However, some co-authors remarked that it is unusual to publish aircraft campaign results without providing the detailed flight paths. For example scientists planning future scientific missions might extract important information from a paper where results are presented together with such campaign details. Inspection of the campaign overview paper by von Hobe et al., (2013) revealed that the flight tracks are there, but not well visible. So in the end we moved Fig. 4 also to the supplement material (see Figure S3 in the Supplement) and indicated the AREA of the flights on the Figures 1 and 3 of the Paper.

And indeed, the pressure level of 30 hPa is above the M-55 flight altitude. The intention to show the temperature map at 30 hPa is rather related to the higher altitudes where large PSC-particles must have been nucleated at first. Then the particles grew for several days until sedimentation moved them to altitudes several kilometers lower prior to the detection at a pressure level of about 50 hPa. This point has been added to the text part, which refers to these figures. We left the 30 hPa contour plots in the paper but in addition supplied the 50 hPa plots in the supplement (Figures S1 and S2 there). The locations of the contours are very similar; only the general temperature level at 50 hPa is approximately 3-5 degrees higher.

Reviewer #1, Specific comment #5: *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.*

Author's Reply to Specific Comment #5: Yes, correct. This paragraph was moved to the observation and results section. Now, first the observation is described, and then the correction explained.

Reviewer #1, Specific comment #6: *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?*

Author's Reply to Specific Comment #6: Following the reviewer's suggestion the statement has been changed for clarification: "In the relevant particle diameter range below 1 μm the Mie curves for the refractive indices of STS and NAT are very close to each other with deviations of less than 10 % between them."

Reviewer #1, Specific comment #7: *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.*

Author's Reply to Specific Comment #7: This part also was moved the "Observation and results" section and completely rewritten with an extra piece of text explaining the origin and meaning of the DoF ratio.

Reviewer #1, Specific comment #8: *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.*

Author's Reply to Specific Comment #8: This analysis is now motivated by referring to the problem of multiple counts in the FSSP-300 data and the potential introduction of other artifacts like shattering.

Reviewer #1, Specific comment #9: *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?*

Author's Reply to Specific Comment #9: A few sentences on the instrument technique were added in the text and some further details in the caption. This "shadow cast" technique is well established in the literature, albeit mostly in connection with larger hydrometeors of 100 μm to 6000 μm in diameter in clouds. So our explanation still is somewhat short, but we hope with the help of the references a reader unfamiliar with the technique will find sufficient detail.

Reviewer #1, Specific comment #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?*

Author's Reply to Specific Comment #10: The MAL-figure illustrates the synoptic nature of the PCSs by means of an instrument which is carried along on board the aircraft (i.e. no distant satellite or fixed ground based remote sensing). Also, without MAL we only could have presented data from a one dimensional, linear, "spaghetti"-line (i.e. lengths of kilometers with a square-millimeters cross section) from the optical particle counters. The miniature lidar adds a two dimensional view of the larger cloud environment.

Especially in the Arctic such synoptic scale PSC of the observed extent are very rare (up to now) and

most publications are concerned with the much smaller lee wave PSCs (i.e. tens of kilometers). Caused by climate change the stratospheric temperatures are decreasing (e.g., Randel et al., JGR, 2009; also referenced now in the paper) and consequently it can be expected that the frequency of occurrence of *synoptic scale* PSCs in the Northern hemisphere will increase, as well as possibly their individual life times. Thus we considered as important to “visually” demonstrate by the in situ instruments, that indeed we sampled the synoptic scale PSCs. Concerning this context we added a sentence in the abstract and the conclusions, as also requested by the second reviewer. Inherently the fact that the lidar figure induced a Reader’s Comment to our submitted manuscript also demonstrates the usefulness of the airborne lidar.

The contours moving with the aircraft only follow the dynamic range and sensitivity limitations of the lidar. Light return signals coming from scatterers too close to the aircraft saturate the detectors. For this reason no data of air closer than 150 - 400m from the aircraft can be recorded. It is like if you had a ground based upward looking lidar mounted on a truck driving up and down mountain roads through a fog. In the newly provided supplement the principle of MAL and MAS are explained, which also includes more detail on the ranges of observability.

Reviewer #1, Specific comment #11: *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.*

Author’s Reply to Specific Comment #11: In the literature (e.g. Pitts et al., 2011) plots of BSR and depolarization have been shown to separate different particle types, with the corresponding ambiguities in the overlapping regions. Here we apply the same principle. The finding that STS particles were present in large number densities is corroborated by the lidar measurements shown in Fig. 13 (i.e. Figure 10 in the revised manuscript) where a scatterplot of aerosol depolarization vs. backscatter ratio is reported. Here the lidar MAL (red crosses) and the backscatter sonde MAS probed the same 20 January PSC (black 15 crosses) as shown in Fig. 13 (i.e. Fig. 10 in the revised manuscript). Also PSC data from 17 January (green squares) and 25 January (blue squares) are included in Fig. 13/10. While the latter show depolarization around 9 %, indicating a detectable presence of aspherical, presumably solid particles, the former two displayed no significant aerosol depolarization, confirming that the STS particle contribution to the optical characteristic of these clouds was dominant. Nevertheless, some signal on 20 Jan. in the depolarization channel is still present and indicates that solid particles, probably NAT, with number densities much below those of the STS droplets, always were present embedded in the predominant STS clouds. In the revised version of the Figure 13/10 we also added the “phase boundaries” between different particle types (i.e. Mix1, STS, Ice,....).

Reviewer #1, Specific comment #12: *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.*

Author’s Reply to Specific Comment #12: The reviewer is right here. We have the problem that these are the only particles collected and brought down to the lab which we have from PSCs. (From our knowledge of the literature -besides Corte et al., 2013,- these might possibly even be the only ones sampled inside Northern hemispheric PSCs.) In other words our data base from the collected particle samples is too small for a separate paper. So we included the results here although this indeed is unrelated to the main focus. In the revised version we gave it its own section title, at the same time also making clear that this is a side note only. Unfortunately we did not operate a counterflow virtual impactor which allows to separate cloud particles from interstitial aerosol particles. For this reason we

can only speculate on the nucleation capabilities of these particles. We added a sentence to this effect in the revised version. At this point we would like to leave the decision to the editor whether this section should remain in the paper or not.

Reviewer #1, Specific comment #13: *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 ($\sim 10 \text{ cm}^{-3}$) and relatively small size ($\sim 1\text{-}1.5$ 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?*

Author's Reply to Specific Comment #13: The high number density of particles in the “mother cloud”, where ice or NAT particles nucleate, does not necessarily have to be maintained while these particles grow and sediment over long distances. The number density is reduced by orders of magnitude due to much higher sedimentation speeds. Moreover, the growth process is highly selective, because particles from the bottom of the cloud which sediment first, deplete the underlying layers for successive particles. This process is well described and modeled in Fueglistaler et al., ACP, 2002. The vertical resolution of the CLaMS model is not sufficient to reproduce this effect in full extent.

Reviewer #1, Specific comment #14: *p.12102, Table 1 – The headings for columns 3 and 4 are identical. Should the heading for column 3 be “ $D > 20$ microns”?*

Author's Reply to Specific Comment #14: Thank you; we corrected this typing error. The other column contains the count results for $D > 15$ microns.

Reviewer #1, Specific comment #15: *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.*

Author's Reply to Specific Comment #15: Yes, good point; we corrected.

Reviewer #1, Specific comment #16: *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?*

Author's Reply to Specific Comment #16: Our copy of the FSSP-300 instrument was used in 1996/7 with “old” analog electronics. Before RECONCILE/ESSENCE this electronics was replaced by modern, fully digital signal processing. (The instrument optics remained the same.) Operating such instruments under these extreme ambient conditions always bears the risk of unnoticed baseline shifts, amplifier settings to drift away from the set-points etc.. Especially at the lower particle size detection limit the instrument is very sensitive to noise and related issues. We were asked (and criticized) at conferences for not proving (outside of the laboratory) that the new electronics indeed gives the same results as the precursor version. For this reason we prepared the data and this figure on the background aerosol because it shows the instrument performances particularly at the small particle size limit. Fortunately in the stratosphere the aerosol variability is mostly caused by major volcanic eruptions. In absence of such eruptions, and after long enough quiescent times, the particle size distributions and the concentration levels should be comparable, even if measured many years apart. This can be also seen from Campbell and Deshler, 2014, and SPARC, 2006. However, since the main reason for the figure is a technical one, we moved it to the supplement.

Reviewer #1, Technical Corrections:

- * p.12074, line 25 – change wording to “: : existence of the most stable hydrate: : :” **Corrected.**
- * p.12074, line 27 – Does “freely floating” mean “individual”? **yes. Changed.**
- * p.12077, line 10 – reword to “: : CDP should have been at a diameter: : :”. **Corrected.**
- * p.12077, line 19 – change “objects” to “particles”. **Changed.**
- * p.12078, line 19 – delete the word “of” after the word “below”. **Corrected.**
- * p.12084, line 28 – reword to “: : particles larger than 15 nm.” **Corrected.**
- * p.12119, line 2 of Fig.16 legend – change “date” to “data”. **Corrected.**

Many thanks to this Reviewer for the careful reading.

Comments of Referee #2: A.R. MacKenzie

Reviewer #2, Specific comment #1: *That said, I would encourage the authors to write a paragraph in the conclusions (and sentence in the abstract) about the likely quantitative impact of this observation on our understanding or modeling of polar ozone. Such a discussion could focus on understanding contemporary processes and/or implications for polar ozone under climate change.*

Author’s Reply to Specific Comment #1: Valid point; we added a sentence in the abstract, and a few more in the introduction/conclusions. This includes three additional references for the decreasing temperatures in the stratosphere as result of climate change.

It seems that even models with non-explicit PSC particle nucleation schemes already reproduce ozone loss quite well, if the relevant parameters are fitted to the current state of the polar stratosphere. Newer models with a temperature dependent NAT nucleation scheme and addition temperature fluctuations along particle trajectories perform even better (Engel et al., 2013). The denitrification by itself is not questioned, and can be measured by remote sensing techniques. Moreover, modeled particle sedimentation speeds can be adjusted by a factor to fit the observations, like in Woiwode et al., 2014.

The observations mainly can serve to improve simulations of the vertical redistribution of trace species by sedimentation. Also for the Arctic polar vortex with more frequent occurrence of lee wave PSCs the particular properties of synoptic scale PSCs have large impact on the ozone depletion from year to year. Therefore, the correct representation of PSC microphysics in large particle size range is relevant. Apparently the occurrence of NAT rocks is a regular feature of synoptic scale PSCs and this point also is highlighted more prominently in the revised version.

Reviewer #2, Specific comment #2: *Although I was able to follow the logic more-or-less, I agree with the other reviewer that, especially for those not in the field, some better sign-posting of the development of the argument would be helpful throughout the manuscript.*

Author’s Reply to Specific Comment #2: The structuring of the text and the order of figures has been changed –and hopefully improved– as also suggested by the anonymous Referee #1.

Reviewer #2: Minor points:

- * Abstract, line 22: spelling of “containing”. **Corrected.**
- * P12076, line 5: *perhaps say “a mass-closure problem”?* **Yes, corrected.**
- * P12085/6. *Could I ask that special care is taken here to make explicit whether diameter or volume is*

being compared when particles are described as “bigger” or “twice as big”. The cubic dependence of volume on diameter obviously makes this distinction important!

Reply: As instance, during the ESSenCe flight the observed volume of the particle phase was twice as big as previously reported. We rephrased a few sentences.

* P12085, line 28: should be “For instance” or “As an example”. **Corrected.**

* P12090: “Thus, one can only speculate...”. **Corrected.**

* P12092. Hypothesised. **Corrected.**

References cited in this Reply:

* Campbell, P., and Deshler, T.: Condensation nuclei measurements in the midlatitude (1982–979 2012) and Antarctic (1986–2010) stratosphere between 20 and 35 km, *Journal of Geophysical Research: Atmospheres*, 119, 2013JD019710, 10.1002/2013jd019710, 2014.

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* Engel, I., Luo, B. P., Pitts, M. C., Poole, L. R., Hoyle, C. R., Grooß, J.-U., Dörnbrack, A., and Peter, T.: Heterogeneous formation of polar stratospheric clouds – Part 2: Nucleation of ice on synoptic scales, *Atmospheric Chemistry and Physics*, 13, 10 769–10 785, <http://dx.doi.org/10.5194/acp-13-10769-2013>, 2013.

* Fueglistaler, S., Luo, B., Voigt, C., Carslaw, K., Peter, T.: NAT-rock formation by mother clouds: a microphysical model study. *Atmospheric Chemistry and Physics* 2(2), 93–98 (2002). DOI 10.5194/acp-2-93-2002, 2002

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* von Hobe et al.: Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions (RECONCILE): activities and results, *Atmospheric Chemistry and Physics*, 13, 9233–9268, <http://dx.doi.org/10.5194/acp-13-9233-2013>, 2013.

* Woiwode, W., Grooß, J.-U., Oelhaf, H., Molleker, S., Borrmann, S., Ebersoldt, A., Frey, W., Gulde, T., Khaykin, S., Maucher, G., Piesch, C., and Orphal, J.: Denitrification by large NAT particles: the impact of reduced settling velocities and hints on particle characteristics, *Atmospheric Chemistry and Physics Discussions*, 14, 5893–5927, <http://dx.doi.org/10.5194/acpd-14-5893-2014>, 2014.