

The authors have replied to reviewer 3's review of the revised-MS (which consisted of 10 further minor revision comments) and have made a series of minor revisions for this 2nd-revised manuscript.

Reviewers 1 and 2 reviewed the revised-MS and ticked the box the study was ready for publication without requiring further edits.

Within this Topical Editor decision, on this 2nd-revised MS, I disclose here that I have published and expert-reviewed PSC papers, and therefore feel qualified to adjudicate on the replies to Reviewer 3's comments.

I have reviewed each of the replies, and the corresponding edits, and in most cases, these are fine and good replies, with this 2nd-revised MS now nearly ready to proceed to publication.

In a few cases however, the revisions made require still some minor corrections.

There are 6 "remaining minor comments" which I enumerate R1 to R6 below.

In 5 of the remaining minor revisions, these relate to the author replies to reviewer comments, and please refer to the supplement PDF, to be able to see the review comments, with having copied-and-pasted the corresponding review-comment in green, and the author-reply in blue.

Once the authors have replied to these remaining comments, and made appropriate further minor edits, I anticipate the 3rd-revised MS will then be able to proceed to publication in GMD.

Line numbers in these remaining minor revision relate to the Track-Changes 2nd-revised MS.

Remaining minor revisions

R1) This 1st of the comments aligns with the 7th of reviewer 3's 10 comments, and consists of two distinct corrections of my own (R1.1 and R1.2), aligned mainly to the modeling specifics.

R1.1) Line 165 – Please add "(from equations 8 and 5)" after "is then calculated" (before "based on.."). (For modelers reading this text re: the sequencing of the processes, it will be helpful to be able to cross-check with the specific physical equations presented in the manuscript.)

R1.2) Lines 186-187 – This sentence seems to be incorrectly formulated, because MERRA is a meteorological re-analysis dataset. I assume the authors meant they used the re-analysis temperature and pressure to calculate the molecular number density, which then is used to calculate the molecular backscatter. It's the 1st stage of this two-stage process that is incorrectly described in the current text. Please re-word the sentence accordingly.

Some readers may wonder why this calculation is not done with the ERA5 meteorological re-analysis, since this is the re-analysis product is used in the nudging system applied in the UKESM1.1 simulations (see line 267 of the TC manuscript).

I get that this is an offline calculation, and then it is a separate decision from that, which re-analysis system to use in this calculation. But maybe add the word "offline" so this is implicit.

R2) This relates to the reviewer 3's comment 2 (in green below). The authors have replied (in blue), and made a generally appropriate minor-edit to the manuscript.

This is mostly fine, but the 2nd bullet in the author replies, although mostly correct, needs to be clear this varying R-ice-NAT boundary-value only became varying since v2 of the CALIOP

(Pitts et al., 2018). Many readers would expect the boundary to be a constant R value, e.g. set at 5.0 for the

CALIOP v1 PSC typing scheme from Pitts et al. (2011), and similar value in the original ground-based lidar classifier scheme of Adriani et al. (2004), on which v1 was based.

There are 2 specific changes here I'm suggesting:

R2.1) Line 201 -- Add to the start of this sentence "Since v2 of the CALIOP particulate-typing scheme (Pitts et al., 2018)...", i.e. "Since v2 of the CALIOP particulate-typing scheme (Pitts et al. 2018), the R_{532} boundary separating enhNAT and NATmix from ice, $R_{\text{NAT|ice}}$ is based on a parameterization of...."

R2.2) Lines 204-205 – The word "daily" here seems anomalous. I think you've used that word to communicate the value is not constant. But better imo to say "the variable RiceNAT value" rather than "the daily RiceNAT value". I couldn't see a reason for the specificity to "daily".

R2.3) Lines 205-206 – The "laid out in" is too colloquial for a peer-reviewed journal article. Suggest to remedy this via re-wording "is from the composition classification laid out in Pitts et al. (2011)" to "is from v1 of the CALIOP particulate classification scheme (Pitts et al., 2011)."

2) Lines 199-200: and Table 1 The boundaries for $R_{\text{nat/ice}}$ is not explained well. The authors claim "The R_{532} boundary separating enhNAT and NATmix from ice, $R_{\text{NAT|ice}}$, is calculated dynamically from coincident cloud-free MLS gas-phase HNO_3 and H_2O data and theoretical R_{532} values for fully developed STS." But this is ambiguous. Is there a maximum for R_{sts} ? If so what is it? How the gas phase concentrations are used in the calculation is not explained. Then some numbers are added to the criteria, 50, 2 in R, and $2.5 \times 10^{-5} \text{ km}^{-2} \text{ sr}^{-1}$ for backscatter, with units that are not backscatter. The source of these numbers is also not explained.

Reply: Thank you for highlighting the ambiguities in the description of the CALIOP classifications. We would like to clarify the following:

- There is no maximum for R_{STS} based on the CALIOP v2 Classification Algorithm documented in Pitts et al. (2018).
- $R_{\text{ice|NAT}}$ is based on a parameterisation of theoretical calculations of R_{532} for fully developed STS over a range of atmospheric pressures and H_2O and HNO_3 concentrations. Therefore, to determine the daily $R_{\text{ice|NAT}}$ values for the CALIOP measurements, daily 'cloud-free' MLS HNO_3 and H_2O concentrations are used.
- The $R > 2$ and $\theta_{\perp} > 2.5 \times 10^{-5} \text{ km}^{-1} \text{ sr}^{-1}$ boundaries for enhNAT are determined from empirical comparisons between CALIOP and MIPAS observations of NAT formed with and without the heterogeneous formation on wave ice.
- The wave ice boundary of $R > 50$ is from the composition classification laid out in Pitts et al. (2011).

To address these ambiguities in the boundaries, we have included the following text in the CALIOP classification description and have corrected the units in the table from $\text{km}^{-2} \text{ sr}^{-1}$ to $\text{km}^{-1} \text{ sr}^{-1}$.

*'... PSCs with $\theta_{\perp}$ exceeding the background threshold are assumed to contain non-spherical (i.e., solid) particles. **The solid-containing PSCs are further classified as either liquid-NAT mixtures (NATmix), enhanced NAT mixtures with $r_{\text{NAT}} \lesssim 3 \mu\text{m}$ and NAT volume density $\gtrsim 1.0 \mu\text{m}^3 \text{ cm}^{-3}$ (enhNAT), ice, or high-number-density ice that forms in mountain waves (wave ice). enhNAT is separated from NATmix using boundaries of $R_{532} > 2$ and $\theta_{\perp} > 2.5 \times 10^{-5} \text{ km}^{-1} \text{ sr}^{-1}$, determined from empirical comparisons between CALIOP and MIPAS observations with and without the NAT band formed heterogeneously from wave ice (Höpfner et al., 2006). The R_{532} boundary separating enhNAT and NATmix from ice, $R_{\text{NAT|ice}}$, is based on a parameterisation of theoretical calculations of R_{532} for ...***

R3) Line 274 – This 3rd remaining revision relates to reviewer 3 querying the precision of the values in Table 2 and lines 270-272. The authors have replied that 3 significant figures is what has been done in other published work using the same NAT scheme. However this study needs to be consistent, across the other values presented, e.g. within Table 3.

R3.1) Firstly, I agree with reviewer 3 values given as 2.47×10^{-3} , 2.47×10^{-3} , 2.47×10^{-2} , is a bit anomalous when most other values are presented at only 1 significant figure.

Please change each "2.47" to "2.5" in Table 3 and lines 176 and 284 (of the Track-Changes manuscript).

R3.2) Please also change "spanning ranges used in other models" to "sampling across the wide range of values used in other models".

The other 2 points I make here (R3.3 and R3.4), are re: the text on line 270-275 re: the measurements from Fahey et al. (2001).

R3.3) On line 273, the text states nNAT values “of the order of $1.0 \times 10^{-5} \text{ cm}^{-3}$ value based on measurements by Fahey et al. (2001)”. The observed value given in Fahey et al. (2001) is $2.4 \times 10^{-4} \text{ cm}^{-3}$ up to $2.0 \times 10^{-3} \text{ cm}^{-3}$. The paper states an uncertainty range of $\pm 30\%$, so although there’s likely spatial variability, still the $1.0 \times 10^{-5} \text{ cm}^{-3}$ value needs to be stated in the context of “a lower limit aligned to measurements of Fahey et al. (2001)” (rather than “based on the measurements of Fahey et al. (2001)”.

R3.4) I noticed an error on line 275, the text says “used to constrain CALIOP observations (Pitts et al., 2011)”. I checked through the Pitts et al. (2011) paper, and don’t see any NAT number concentration being used there. It’s fine to say those values were used simply from the range observed in Fahey et al. (2001), it doesn’t need any additional justification. Suggest to delete the “and used to constrain CALIOP observations (Pitts et al., 2011)”.

4. Lines 270-272 and Table 2: Why are these numbers three significant figures. That is very exact for an assumed number that can vary by three orders of magnitude.

Reply: Thank you for your comment. We chose three significant figures as this is what has been done in other published work using the same NAT scheme (van den Broek et al. 2004, Kirner et al. 2011, Weimer et al. 2021).

R4) Line 299 of Track-Changes MS → “... their relative abundance is comparable” isn’t quite right. I think maybe you meant that comparing the two qualitatively is still a good test of the model, or so. Suggest to change “their relative abundance is comparable” to “they can be compared qualitatively”.

5. Lines 291-293: The authors should briefly explain why the counts of observations cannot be directly compared?

Reply: Thanks for raising this point. We cannot compare the counts of observations with the grid-cells for the simulations because they are on different spatial scales. CALIOP’s counts are based on data at a horizontal resolution of 5 km, whereas the model simulation’s output are at a coarser resolution of ~ 130 km in the latitudinal direction. We have included the following to clarify this:

*‘Note that while the values of the counts of observations for CALIOP and grid-cells for the simulations in the 2D histograms cannot be directly compared due to **their different spatial resolutions, their relative abundance is comparable.**’*

R5) Line 304 -- This is the text added from Reviewer 3’s 6th comment (copied & pasted in green below). The comment is “Since no ice particles appear in the Control run how can they produce an STS signal”.

The authors’ edit, to add “with low beta_perp signals” does not really help, as both NAT and ice particles are non-spherical, and thus must implicitly have a significant beta_perp signal.

I’m not sure if the authors maybe meant that this instance is from the model predicting only very low number concentrations of these NAT and ice particles?

Suggest to maybe change “with low beta_perp signals” instead to “at very low number concentrations”.

6. Lines 297: Since no ice particles appear in the Control run, how can they produce an STS signal?

Reply: Thank you for this clarification. The CONTROL does produce ice, but due to the low number density and surface area density of these ice particles, there are no points that classify as ice using the CALIOP classification based on optical parameters. We have clarified this in line 297 which is now:

*‘(Note that since CONTROL includes no STS particles, the STS optical signal likely arises from NAT and ice particles **with low $\beta_{\perp}$ signals.**)’*

R6) This comment relates to the 8th of Reviewer 3’s comments, and the reviewer has made a mistake here, in saying “...the boundaries between the various PSC types are controlled by Eqs. 14 and 15”.

That’s clearly not correct, as equations 14 and 15 don’t set the thresholds, rather they set the actual values of the perpendicular backscatter, and the backscatter ratio (β and R).

I think the reviewer is referring to the fact that the boundaries between ice and NAT are no longer at a fixed value of the backscatter ratio (it was set at $R=5.0$ at v1 of the CALIOP algorithm, in Pitts et al., 2011).

Unintentionally, I think the reviewer's comment highlights that Pitts et al. (2018) do not seem to have included any equation for this now varying NAT-ice boundary. They explain this is a "parameterization of theoretical calculations of R_{532} for fully developed STS", but no equation is given.

And then I'm wondering how the authors did actually calculate this for the methods stated here? Does ICON-ART already include some lines of code for this parameterization referred to here? Or did co-author Mike Pitts provide some code for the authors to generate the boundary values?

Please can the authors reply to clarify how this was calculated, and state in section 2.1.1 (or similar) how this was actually calculated for post-processing to the model values shown in Figure 1d.

The edit, refers to threshold being determined by Equations 14 and 15, but as above, that's not correct.

Maybe best to change "(including measurements uncertainty) are determined by Eqns. 14 and 15" instead to "(including measurements uncertainty from equations 16 and 17)" or similar re-wording. Or alternatively just delete "thresholds" and change the brackets there to commas, with changing "and therefore shift based on..." instead to "with thresholds varying based on..."?

8. Lines 290..., Figs. 1 and 2: The authors should remind the readers that the boundaries between the various PSC types are controlled by Eqs. 14 and 15 which are likewise dependent on the median values plus one median deviation in $\beta_{\perp}$ and scattering ratio. Thus, the boundaries will shift based on the measured and modeled median values. While this characteristic adds to the complications of the measurement to model comparison, it is also an indication of the agreement/disagreement of model with measurement. This latter point perhaps should be mentioned in the panel to panel discussion.

Reply: Thank you for highlighting this. We agree this is important to address and have included the following two sentences near the end of Section 3.1 (Lines 332 – 335):

'Note that the mean $\beta_{\perp}$ and R_{532} thresholds (including measurement uncertainty) are determined by Eqns. 14 and 15 and therefore shift based on the measured and modelled values. In Fig. 2, the mean $\beta_{\perp}$ line is lower across all periods and simulations compared to CALIOP, indicating the generally lower $\beta_{\perp}$ values in the distribution of model optical values compared to CALIOP.'