

Dear Graham,

We are extremely grateful for your careful review of the revised manuscript. Your comments and suggestions have improved the manuscript's clarity, which we are very appreciative of. We have carefully addressed all of your comments and are pleased to submit a new revised version of our manuscript.

Please find the point-by-point response to your comments. As with the previous responses, our responses are in blue and new text in the manuscript are **bold** and *italicised* text.

Sincerely,

Isabelle Sangha, on behalf of all the coauthors

Topic Editor Comments

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.)

Reply: Thank you for suggesting this. We have followed your suggestion and included the reference to the equations. The sentence is now:

‘The formation, growth, or evaporation of NAT is then calculated (from Eqns. 8 and 5) based on the remaining gas-phase HNO_3 .’

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.

Reply: Thank you for this clarification. We have followed your suggestion and changed the sentence to clearly state that the reanalysis temperature and pressure fields from MERRA-2 are used to calculate the molecular number density which is then used to determine the backscatter. The sentence is now:

‘The molecular number density (calculated using temperature and pressure fields from Modern-Era Retrospective analysis for Research and Applications, version 2 (Gelaro et al., 2017) reanalysis) is used in the theoretical calculation described in Hostetler et al. (2006)...’

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.

Reply: We agree it is useful to clarify the optical properties are calculated offline for UKESM1.1 and have followed your suggestion and added the word ‘offline’ at the beginning of section 2.3.1, when describing how optical properties are calculated from UKESM1.1 output (see italicized text below). However, we would like to highlight that the MERRA-2 reanalysis is used in calculating the molecular backscatter for the CALIOP observations (described it Pitts et al. (2018)). The optical properties for UKESM1.1 are calculated using Mie theory and T-matrix calculations in the Zurich Optical Microphysical Model (ZOMM).

New text in Section 2.3.1: *‘To compare PSCs simulated by UKESM1.1 with those observed by CALIOP, the particle size distributions from the model are used to calculate optical parameters **offline** as they would be observed ...’*

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 R532 boundary separating enhNAT and NATmix from ice, RNAT,ice is based on a parameterization of....”

Reply: Thank you for this comment. We have included your suggestion, however we use the wording ‘classification algorithm’ to remain consistent with the rest of the manuscript. The sentence now reads:

‘Since v2 of the CALIOP classification algorithm (Pitts et al., 2018), the R_{532} boundary separating enhNAT and NATmix from ice...’

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”.

Reply: We agree that the word ‘daily’ here isn’t the best way to communicate that the $R_{\text{NAT|ice}}$ varies. Following R6 below, this sentence has been rephrased and no longer includes ‘daily $R_{\text{ice|NAT}}$ ’. Please see R6 for the new wording for this sentence.

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).”

Reply: Thank you for suggesting a rewording. We agree and have made the suggested change to the sentence, but we use the wording ‘classification algorithm’ to remain consistent with the rest of the manuscript. The sentence now reads:

‘The wave ice boundary, $R_{532} > 50$, is from the v1 CALIOP classification algorithm (Pitts et al., 2011).’

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).

Reply: We have changed the NAT number densities in Table 3, and references in the text, from values of ‘2.47’ to ‘2.5’ in accordance with this suggestion.

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

Reply: Thank you. Following this suggestion we have amended the sentence to read:

*‘For ice PSCs, where observations are limited, n_{ice} was set to 0.001, 0.01, 0.1, and 0.25 cm^{-3} , and 0.25 cm^{-3} , **sampling across the wide range of values used in other models** (Tritscher et al., 2019; Steiner et al., 2021; Weimer et al., 2021; Zhu et al., 2017), as well as the ice number densities considered by Pitts et al.(2018).’*

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 +/- 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)”).

Reply: Thank you for highlighting this. We would like to clarify that this is referring to the order of magnitude of the values used for $n_{\text{NAT,lim,b}}$ which results in $n_{\text{NAT,max}}$ of 10^{-4} cm^{-3} in agreement with the Fahey et al. (2001) measurements. However, we agree that this could be explained in a clearer way, and so have amended the following sentences:

'In the implementation of the same kinetic NAT parameterisation used here, Kirner et al. (2011) and Weimer (2019) use $n_{\text{NAT},\text{lim},b}$ values of the order of 10^{-5} cm^{-3} , resulting in a total maximum number density limit, $n_{\text{NAT},\text{max}} = \sum_b n_{\text{NAT},\text{lim},b}$, of the order of 10^{-4} cm^{-3} based on measurements of Fahey et al. (2001). Here, we additionally investigate the effect of increasing the $n_{\text{NAT},\text{lim},b}$ values to the order of 10^{-4} and 10^{-3} cm^{-3} to cover the range of number densities observed (Fahey et al., 2001; Biele et al., 2001) and used to constrain CALIOP observations (Pitts et al., 2018). The size bins and different orders of magnitudes used for the $n_{\text{NAT},\text{lim},b}$ are summarized in Table 2 and result in values of $n_{\text{NAT},\text{max}}$ equal to $2.5 \times 10^{-2} \text{ cm}^{-3}$, $2.5 \times 10^{-3} \text{ cm}^{-3}$, and $2.5 \times 10^{-4} \text{ cm}^{-3}$ (for $n_{\text{NAT},\text{lim},b}$ values of the order of 10^{-3} , 10^{-4} , and 10^{-5} cm^{-3} , respectively).'

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)”.

Reply: Thank you for this comment. In reviewing the line, we realised the NAT constraints are defined in Pitts et al. (2018), not Pitts et al., (2011). We have corrected this reference so the sentence now reads:

'Here, we additionally investigate the effect of increasing the $n_{\text{NAT},\text{lim},b}$ values to the order of 10^{-4} and 10^{-3} cm^{-3} to cover the range of number densities observed (Fahey et al., 2001; Biele et al., 2001) and used to constrain CALIOP observations (Pitts et al., 2018).'

R4) Line 299 of Track-Changes MS à “... 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”.

Reply: We have followed your suggestion and have changed the wording in the sentence which is now:

*'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 can be compared qualitatively.***

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”.

Reply: Thank you for this suggestion. We have followed your advice and changed the wording of this sentence, however, we have used ‘number densities’ to remain consistent with the rest of the manuscript. The new sentence reads:

*'(Note that since CONTROL includes no STS particles, the STS optical signal likely arises from NAT and ice with **very low number densities.**)'*

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 (b 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 R532 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..."?

Reply: Thank you for raising these points. Regarding the confusion around the 8th of Reviewer 3's comment about Eqns. 14 and 15, we would like to confirm that these are the correct equations, and they refer to the boundaries that $\beta_{\perp}$ and R_{532} must exceed to be defined as a PSC occurrence. However, to make this clearer in the manuscript we have added the following to the sentence referring to Eqns. 14 and 15:

*'Note that the mean $\beta_{\perp}$ and R_{532} thresholds (including measurement uncertainty) are determined by the **boundaries defined** in Eqns. 14 and 15 and therefore shift based on the measured and modelled values'*

Concerning the second point raised in this comment about the equation for the varying NAT-ice boundary from Pitts et al. (2018), we would like to provide here further explanation for this calculation. The values for $R_{ice|NAT}$ derived by Lamont Poole for CALIOP are based on daily MLS HNO_3 and H_2O values computed for 'cloud-free' conditions in equivalent latitude bands and potential temperature layers ($h2oeq$ and $hno3eq$ in the equations below, in ppbv and ppmv, respectively). $R_{ice|NAT}$ has dimensions (neql, nd, npt), where:

- neql = the number of equivalent latitude bands: 80-90 S, 70-80 S, and 60-70 S (opposite for NH)
- nd = number of days in MLS cloud-free files: weekly values (mid-week)
- npt = number of overlapping 100 K thick potential temperature layers: 9; 250-350 K, 300 – 400 K, 350 – 450 K, 400 – 500 K, 450 – 550 K, 500 – 600 K, 550 – 650 K, 600 – 700 K, 650 – 750 K

For a given equivalent latitude band (ieql), day (id), and potential temperature layer (ipt), $R_{ice|NAT}$ is calculated using the following derived relationship:

$$a_1 = 0.398 - 0.00293 * h2oeq(ieql, id, ipt),$$

$$a_2 = -0.00575 + 9.33e-4 * h2oeq(ieql, id, ipt),$$

$$R_{ice|NAT}(ieql, id, ipt) = 1 + a_1 * hno3eq(ieql, id, ipt) + a_2 * (hno3eq(ieql, id, ipt))^2,$$

This $R_{ice|NAT}$ data is used for PSC detection and composition classification for both CALIOP and the model optical data. The $R_{ice|NAT}$ for a specific CALIOP observation, or model grid-cell is estimated by finding the appropriate equivalent latitude band and potential temperature layer that the observation or model grid-cell falls into and use the corresponding weekly $R_{ice|NAT}$ value.

We feel the inclusion of this relationship is too detailed for this manuscript as it is related to the CALIOP classification algorithm, rather than the model implementation. However, we have changed the following in the manuscript to further clarify how the values are estimated for CALIOP observations and model grid-cells.

In Section 2.2 :

*'Since v2 of the CALIOP classification algorithm (Pitts et al., 2018), the R_{532} boundary separating $enhNAT$ and $NATmix$ from ice, $R_{NAT|ice}$, **is derived from relationships based on MLS HNO_3 and H_2O***

concentrations computed for weekly ‘cloud-free’ conditions in equivalent latitude bands and potential temperature layers. The $R_{\text{NAT|ice}}$ for a specific CALIOP observation is then estimated by finding the appropriate equivalent latitude band and potential temperature and using the corresponding $R_{\text{NAT|ice}}$ value for the week.’

And in Section 2.3.1:

‘Additionally, the $R_{\text{NAT|ice}}$ value is estimated for each grid-cell using the same data and method as CALIOP (described above).’