Reply to the comments of reviewer 1 on the manuscript

Radiative transfer simulations and observations of infrared spectra in the presence of polar stratospheric clouds: Detection and discrimination of cloud types

by C.Kalicinsky et al.

We thank the reviewer for the helpful comments and recommendations. In the following, we discuss the issues addressed by the reviewers and explain our opinions and the modifications of our manuscript.
We enumerate the comments and repeat them in bold face. The modifications of the manuscript are displayed in the marked-up manuscript version as colored text. Deleted parts are shown in red and new or modified text parts in blue.

1 Comments

General comments
Based on radiative transfer simulations, the authors present a method to detect and discriminate PSCs in airborne CRISTA-NF observations. NAT and ice are identified by using their characteristic spectral patterns. Detected PSCs, which do not show these signatures, are classified as STS. Assuming spectral characteristics of spherical particles, NAT PSCs are sub-grouped into small NAT (median r≤1.0μm), medium-sized NAT (median r=1.5μm – 4.0μm) and large NAT (median r≥3.5μm). Furthermore, a new method to detect the bottom altitude of non-opaque clouds is introduced. Application of the proposed methods to CRISTA-NF data is presented for one flight during the RECONCILE field campaign.
The presented study is interesting for PSC-related research, since aspects of PSC detection and classification are investigated in further detail and extended. The CRISTA-NF observations provide another interesting data set in the winter 2009/10, which was characterised by wide-spread PSC occurrence. The proposed approach to distinguish between different size classes of NAT particles might have potential to extend PSC classification using satellite instruments. However, the presented size classification is elaborated using model simulations only. Direct and detailed comparisons with observed spectra are not presented. Only spherical NAT particles are taken into account, while previous studies suggest that particle shape is an important issue. Comparisons with in situ observations could be helpful to test the proposed method. The method to determine cloud bottoms has the potential to gain more information on PSCs and thin cirrus clouds in the vertical domain. From my point of view, the manuscript is interesting and should be considered for publication in AMT after the following points have been addressed.
Main points:

1. **From my point of view, it seems that the presented PSC classification and the size classification of NAT particles are elaborated separately from the observations. Simulated spectra in Figures 1 and 5 are presented in arbitrary**

units and are scaled individually, while only a few observations are shown separately in Figure 11 using radiance units. Furthermore, only a part of the used spectral range is shown for the observations (it seems that LRS 5 data is not shown). Therefore, the presented plots do not allow to evaluate whether the simulations reproduce details of the observations. From my point of view, this aspect is important, especially if issues related to particle shape are addressed. Overlays or residuals between simulated spectra and observations would allow to evaluate to which degree the underlying assumptions of the classification are supported.

It is correct, our classification method is derived from simulated NAT, STS, and ice PSC spectra. Our intention was to use a wide spread of different situations to stake out the possible range of signals due to variations in the PSC particle size distribution (as done before by Höpfner et al (2006), Spang et al. (2012,2016)). As our detection and classification method is based on relative effects, i.e. colour ratios and brightness temperature differences, we can show where the CRISTA-NF measurements are located in the color ratio and BTD range, without fitting the absolute radiance of each single spectrum. Scaling the spectra to 1 at $833\,\mathrm{cm}^{-1}$ (Figs. 1,2,5) helps to point out the relative differences that our method relies on.

We added the second CRISTA-NF channel. We included new figures in the manuscript that show the correlations between the NAT indices/ the BTD and the CI for the observations of CRISTA-NF (Fig. 9) to allow for a comparison with the simulation results (Figs. 3,6). We also included data from additional flights of the RECONCILE campaign to demonstrate that the broad range of simulations covers well the range of observations (Fig. 9).

All results are shown in the new subsection 4.1.

2. **The proposed size classification is based on spectral characteristics of spherical NAT particles. However, field observations (Molleker et al. 2014) and laboratory experiments (Grothe et al., 2006) support a highly aspherical particle shape of large NAT particles or alternative habits. In a well-constrained scenario, the observed spectral fingerprint of large NAT particles has been shown to be clearly better compatible with highly aspherical particles (Woiwode et al., 2016). From my point of view, potential uncertainties of the proposed size classification related to the adopted particle shape should be discussed.**

   The study is based on simulations for spherical particles as this particle shape can be simulated with less computational effort compared to aspherical particles, which is an important factor with respect to the large number of simulations that were carried out. We did not question the existence of aspherical NAT. However, our simulation results demonstrate that spherical particles can produce all the different appearances of the NAT feature (typical feature, shifted feature, step-like behaviour) including those previously attributed to highly aspherical NAT. We think that this is an important point to consider as it implies that IR limb emission measurements alone are not sufficient to derive the existence of aspherical NAT, in particular for cases without additional information on the atmospheric state and cloud particle size distribution.

   Our approach of grouping the NAT particles into 3 size bins based on the spectral signature we do not consider affected by the particle shape for the following reasons. The

small particles (< 3 μm) are considered not or only slightly aspherical. In our study we used the term 'small' for particle size distributions with median radii ≤ 1.0 μm. Also Woiwode et al. (2019) use the method by Höpfner et al. (2006), which is based on simulations using spherical particles, to detect these small particles. A substantial fraction of our 'medium' size group (median radii $\leq 4\,\mu$m) can also be considered nearly spherical. The shifted NAT signature reported by Woiwode et al. (2016) for highly ashperical NAT is bi-modal with median radii of 2 and 4.8 μm, where the latter is dominating the signal. This means that the shifted NAT signature that so far was solely attributed to the asphericity, has only been observed in the presence of larger NAT particles. Further, Woiwode et al. (2019, Fig. 1) show that for median radii ≥ 5.0 μm a step-like NAT-feature can be observed. Hence, our result that the shift of the NAT signature is associated with NAT particle size is in line with the results of Woiwode et al. (2016, 2019). Due to particle asphericity our size bin boundary between 'medium' and 'large' has uncertainties. We added these considerations and the contraint that our size bin boundaries are only valid under the assumption of spherical particles to the discussion.
A detailed comparison between the effects of aspherical, mono-modal , and bi-modal spherical cases is beyond the scope of the paper and should be subject of a future study.

3. **The spectral range used by the windows for NAT index-1 to -3 focuses only on the spectral region around the feature at $\sim$820 cm^{-1}. In previous work, it has been shown that, beside details of the spectral feature at $\sim$820 cm^{-1}, the overall spectrum and especially the spectral region towards 960 cm^{-1} respond to particle size and shape (Woiwode et al., 2016). Are there certain (e.g. instrument-related) reasons for not considering the full available spectral range (and particularly the region 960 cm^{-1}) to constrain the size classification and define the criteria?**
Our detection method is able to detect nearly all simulated NAT scenarios by using the spectral range between 810 and 834 cm^{-1}. It misses only a small part of the very large particles with radii of 8 μm (see Fig. 4 b)). The gradient between 834 cm^{-1} and 950 cm^{-1} is similar for NAT and STS spectra as shown in Fig. 6. We used 950 cm^{-1} instead of 960 cm^{-1} to avoid CRISTA-NF measurement artefacts at the boundary of this channel, but checked in the simulations that both windows deliver similar results. Thus, adding the 950 cm^{-1} spectral region to the classification does not improve the discrimination between NAT and STS.
Moreover, the simulations for NAT/STS mixtures and bi-modal NAT showed that the region around 950 cm^{-1} is very sensitive to these combinations (see Fig. 5). In our opinion there are multiple ways to obtain similar radiance values in this region. By contrast to this, the spectral feature in the region 810 – 820 cm^{-1} always gives information on the particle type and size, even in the case of the mixtures.

4. **Previous studies showed the spectra of large NAT particles respond significantly to variable radiation from the troposphere/surface, which is scattered by the large particles into the line-of-sight (see Woiwode et al., 2016, 2019). The signal is modulated by the absence or presence of tropospheric clouds. To me it is not clear whether this aspect has been taken into account and which uncertainties might result in the size classification. (Were tropo-**

spheric clouds considered for the discussed flight?

The influence of tropospheric clouds was intensively studied during the analysis of the radiative transfer simulations for various PSC situations for the MIPAS-Env satellite instrument (Spang et al., 2012, 2016) and the results can be transfered to here. A tropospheric cloud below the PSC mainly influences the absolute radiance values where the cold tropospheric cloud leads to lower overall radiance values. The appearance of the spectral signature (typical peak, shifted peak, or step-like behaviour) in the region 810 cm^{-1} to 820 cm^{-1} is nearly unchanged. Simulations by Woiwode et al. (2019) also show this. As our detection method is based on colour ratios, i.e. relative effects, the change of the absolute radiance values does not significantly influence the analysis. Especially in the case of STS or ice, which define the separation lines (see Fig. 3), the data points in the scatter plots only slightly shift along the correlation lines/regions for the specific PSC type. Thus, the separation lines are still valid in such cases.
We added a new subsection 3.2.2 to the manuscript.

5. **Application of the proposed classification scheme is presented for one flight and comparisons with in situ observations are not provided. In situ observations during the same flight suggest particle radii exceeding 7.5 and 10μm (see Molleker et al., 2014,Table 1), while the proposed size classification suggests STS and medium-sized NAT (r=1.5μm – 4.0μm) here. Are these results compatible? A comparison with in situ observations and including further flights could be helpful to evaluate (and potentially optimise) the proposed classification scheme.**
   We applied the proposed classification scheme to all PSC flights (1-5) during RECONCILE. The results are now presented in Figure 9 and discussed in Section 4.1.
   Unfortunatelly, the number of particles with radii $> 7.5\mu$m (59 particles in the whole flight) is the only information about the size distribution during flight 3 (22.01.2010). Without any relation to the number of small particles a serious comment on the agreement between CRISTA-NF and those measurements cannot be made. Grooß et al. (2014) present a size distribution for a part of this flight, which shows that most particle radii lie in the range < about 6 μm with a maximum at about 2.5/3 μm. This agrees with our results. The same is true for flight 4 (shown in Molleker et al. (2014)). However, we refrain from such a simple comparison between in situ observations and our results as there are too many differences between the instruments. Since CRISTA-NF observes all air masses along the line of sight (up to hundreds kilometres away from the aircraft), the air masses detected by the in situ instruments may play only a minor role for the infrared spectra.

6. **Regarding the detection method for cloud bottoms, it would be interesting if the authors could discuss how the method responds if the cloud bottom is located above flight altitude and in the presence of several PSC layers located above each other. Such scenarios are frequently found in polar winters (e.g. Pitts et al., 2018).**
   When the PSC is located above the flight altitude the CI typically increases with decreasing altitude from flight altitude downwards, because the length of the line of sight inside the PSC decreases also. Thus, there is not the typical large increase of the CI indicated by the minimum of the CI gradient when leaving the cloud. Both, the CI

and the CI gradient, typically show the lowest values at the points closest to the flight altitude.
In the case of layered PSCs, where PSCs of different type are lying above each other, the results depend on the optical thickness of the different layers. On the one hand, the CI gradient minimum is still located at the bottom of the complete PSC, when the lower layer is optically thick enough and the transition to the cloud free atmosphere shows the largest increase in CI. On the other hand, when the lower layer is optically very thin, it can occur that the CI gradient minimum is located directly below the upper layer. In this case the transition from the upper layer to the lower layer leads to a larger increase in CI than the transition from the lower layer to the cloud free atmosphere.
We added these information to section 3.5.

Specific comments:

1. **L15, L492 It should be mentioned that the size classification assumes spherical NAT particles.**
   We added the information that the classification is based on spherical particles.

2. **L35 References to further studies focusing on model simulations and comparisons with observations might be considered (e.g. Zhu et al., 2015; Zhu et al., 2017; Khosrawi etal., 2017, Tritscher et al., 2019).**
   We added additional information. See Reviewer 2.

3. **L39ff Further PSC detection techniques (e.g. lidar, in situ) might be mentioned (e.g. Achtert and Tetsche, 2014; Molleker et al., 2014; Pitts et al., 2018).**
   We added information on other measurement techniques.

4. **L51 It should be noted that relatively high volume densities are required for this signature (see Höpfner et al., 2006, 2006b), which are found less frequently in the Arctic.**
   We added this information.

5. **L67, L227 "Very small" should be defined.**
   The sizes that are necessary to shift the spectral feature are evaluated in this paper and the numbers are given later in Sec. 3. Thus, we do not add information in the introduction.
   In the case of the BTD only the particles $< 1.0\ \mu$m lead to a large radiance decrease between the two micro windows. We added this information.

6. **L67 "Another spectral feature" should be explained. To me it seems to be the same spectral signature of beta-NAT particles reported by previous studies, which is modulated by the actual PSC scenario, particle size distribution, particle shape and scattered light from below. "A similar spectral feature" seems more appropriate to me.**
   It is a spectral signature of beta-NAT, but in the first cases it is a peak and here a step-like feature. Thus, it is another appearance of the beta-NAT signature. We corrected this.

7. **L70 It should be mentioned that also other particle modes between 1μm and 6μm were analysed by the same study (see Woiwode et al., 2016, Table 2, Fig. 10ff, Appendix B).**
   We added this information.

8. **L72 It should be mentioned that particle shape and scattered radiation from below where also found to be important parameters by the same study.**
   We added this information.

9. **L127ff Information on the along-track sampling would be helpful.**
   We added this information to Sect. 2.1 as this section describes the CRISTA-NF instrument.

10. **L167 The overall cloud scenario used is well-described. However, the tropospheric cloud scenario and implications for scattered radiation from below, which is scattered by the particles, are not addressed (see 4)).**
    See general comments 4.

11. **L163 Did the simulations account for CFC-12? (spectral band centered at ~920 cm^{-1}).**
    No, because the influence of CFC-12 at 940 cm^{-1} or larger wave numbers is insignificant.

12. **L202, L222 Detailed analysis in the Woiwode et al. (2016) showed that the shape of the feature is sensitive to particle shape.**
    As these sentences only refer to our simulations and explain the observed results, we find that the sentences are correct as they are.

13. **L230ff See 1)-4): The observed spectral fingerprint is not exploited fully to define of criteria of the classification. E.g. the spectral region around 960 cm^{-1}, which was shown to respond significantly to size distributions and particle shape (see Woiwode et al., 2016) is not considered. It is not clear to me whether modulation of the spectra by scattered light from the below is considered. Direct comparisons between observed and simulated spectra are not presented to verify the underlying assumptions.**
    See general comments 1-4.

14. **L279, L314, L336, L507 It should be noted that these statements on the performance are valid within the assumptions made. Verification by direct comparisons of simulated and observed spectra and comparisons with other observations are not provided here.**
    As the whole Section 3 describes our simulations and the derived results, the statements are given with respect to the simulations. Nonetheless, we added some additional information on that.
    In L507 the statement is made with respect to the observations, but as we showed spectra showing the shifted feature and, additionally, added comparisons between simulations and observations (see general comments 1), we think the statement is correct here.

15. **L315ff For comparisons with other studies, it would be helpful to include a Figure showing the used size distributions.**

As the number of scenarios of bi-modal NAT particle size distributions is 115, the number of size distributions is also. We think that this is too much to show all size distributions. Furthermore, all relevant parameters of the size distributions are given in Tab. 3.

16. **L383ff See 6): It would be interesting if the authors could discuss how the method responds if the cloud bottom is located above flight altitude and in the presence of several PSC layers located above each other. Such conditions are frequently found inpolar winters (e.g. Pitts et al., 2018).**
See general comments 6 and section 3.5.

17. **L392 The motivation for choosing the discussed flight should be provided.**
We also included results for other flights (see general comments 1). We show more details for flight 3 as it shows the largest NAT signal of all flights and gives a good example. This information is added.

18. **L397 It would be helpful to indicate the location of the selected two profiles in Fig. 10 and comment on the composition derived in Fig. 10b.**
We added arrows to the Fig 10 a) and b), which are now Fig. 11 b) and c).

19. **L416ff See 5): Considering one case study constitutes only a limited test for a new classification scheme. Further RECONCILE PSC flights, collocated in situ observations and e.g. CALIPSO observations could be used to test (and potentially optimise) the proposed classification scheme.**
We added additional flights (see general comments 1).

20. **L422 Why are only spectra with CI $<$ 3.0 shown here? (compare Fig. 3 and L388)**
The separation between the different types of PSCs and also for the different NAT sizes diminishes with increasing CI. This can be seen in Fig. 4. When all spectra with CI $<$ 5 are considered more of the large particles cannot be detected and also medium size particles are missing. Furthermore, the separation between the different size regimes is not so distinct. Thus it is advisable to use only spectra with CI $<$ 3, as these problems are largely reduced then. We added information to Sect. 3.2.1.

21. **L424 See 1): Figure 11 shows examples of PSC spectra in radiance units, while Figures 1 and 5 show simulations in arbitrary units, which are individually scaled. Clear comparisons between simulations and observations are not possible. Only a part of the used spectral range is shown for the observations (compare L99). Overlays or residuals between simulated and observed spectra using the same units would be helpful to evaluate to which degree the simulations meet details of the observations.**
See general comment 1.

22. **L435,436 See 5): Here, the CRISTA-NF observations are classified as STS or medium sized (median r=1.5-4.0μm) NAT. However, collocated in situ observations suggest particles with radii exceeding 7.5 and 10μm (see Molleker et al., 2014, Table 1). Are these results compatible? It would be interesting to compare size distributions, which are supported by the CRISTA-NF**

simulations, with the in situ observations.
See general comment 5.

23. **L472 The conclusion on the spectral range from 833 to 960 cm^{-1} is not supported sufficiently by Fig. 5, since the spectral range from 840 to 940 cm-1 is not shown.**
In our opinion it is supported. Both spectral ranges are shown and there is no difference between the two spectral ranges in relative as well as in absolute values. Nonetheless, we rephrased the sentence and used the term difference.

24. **L474ff From my point of view, the conclusion "very similar" is not sufficiently supported. The Woiwode et al. (2016,2019) studies used a broader spectral range, higher spectral resolution and spectra in absolute radiance units (which constitutes another "piece of information"). Furthermore, these studies used direct overlays and residuals of simulated observed spectra to analyse the spectral fingerprint of large NAT particles in detail and to define criteria for detection. In the study presented here, the full available spectral range is not exploited and no direct comparisons between simulated and observed spectra are provided. However, within the above mentioned limitations, the conclusion on similarity is supported to some degree by the Woiwode et al. (2016) study, Appendix B, for the small and medium particle sizes. In this study it was shown that for small NAT particles (r=1μm), spherical and aspherical particle populations show an almost identical spectrum, while at r=3μm the signatures start to diverge notably. Furthermore, one might speculate that particles become gradually more aspheric when growing to large sizes, and that (nearly) spherical particles are a suitable assumption at earlier growing stages.**
It is correct that Woiwode et al. (2019) used a wider spectral range, a better resolution etc. for their simulations. Thus, a direct comparison between the two simulations is not advisable. However, as the simulation for the bimodal NAT size distribution shows an enhanced step from 818 to 833 cm^{-1} and no difference between the spectral regions 833 to 960 cm^{-1}, in our opinion the spectrum will fullfil all criteria of the described detection method. For this "comparison" the whole spectrum is not necessary and we simulated all spectral windows used by Woiwode et al. (2019) algorithm. Unfortunately, a direct use of this method is not possible as the absolute radiance values of the two instruments, CRISTA-NF and MIPAS-Envisat, are different (see specific comments 27.).
We rephrased the sentence accordingly.

25. **L476ff It has been shown before that a similar signature can be simulated by assuming spherical particles (see Woiwode et al., 2016, Appendix B). Furthermore it should be noted that the possibility of some variability in the NAT phase regarding particle habits has been clearly mentioned in previous studies (Molleker et al., 2014; Woiwode et al.,2016) and has not been ruled out in the Woiwode et al. (2019) study. However, using the combination of a wide spectral range, high spectral resolution, and supporting information from in situ observations, the Woiwode et al. (2016,2019) studies were not successful in reproducing details of developed spectral fingerprints of large NAT particles by assuming spherical particles. The observed combination of**

a strong “step-like” feature and a flat spectral baseline of the observed spectra towards higher wave numbers (described by the simplified “hockey-stick” picture in Woiwode et al.,2019) could not be reproduced. Close inspection of the spectra showed that Mie simulations of spherical particles always showed significant differences from the observed ”shifted feature” around 820 cm^{-1} and a significant negative slope and/or an “upwardarching” of the spectral baseline at higher wave numbers, and further discrepancies. These discrepancies resulted in significant patterns in the residuals. Using spectral characteristics of highly aspherical particles clearly improved the residuals. Thus, the spectrum clearly includes information on particle shape. Also from other work it is known that infrared spectroscopy allows to infer information on particle shape (Wagner et al., 2005). However, it is understandable to me that within the limited spectral range considered here and without detailed comparisons between observations and simulations a clear decision appears to be not feasible here. From my point of view, the question is not only whether any “hockey-stick” signature can be modelled, but also whether details of the entire accessible spectral fingerprint in developed signatures of large NAT particles can be reproduced, and how the results compare with other observations.

Although our simulations as well as the CRISTA-NF measurements include the three spectral windows, here we consider our 950 cm^{-1} a sufficiently good substitute for the MIPAS 960 cm^{-1} window, proposed by Woiwode et al. (2019) to identify aspherical NAT, we cannot apply the method as it relies on absolute radiances that are inherently different for space-based MIPAS and air-borne CRISTA-NF measurements. However, assuming spherical NAT particles and a bi-modal size distribution our simulated spectra qualitatively reproduce the spectral signature attributed to aspherical particles. To our knowledge Woiwode et al. (2019) did not investigate the effect of bi-modal size distributions on the spectra and hence, we think this should not be ruled out. We rephrased the sentence here.

26. **L477f This sentence should be revisited, since this has been done in before (Woiwode et al., 2016,2019). Of course, any further case studies including in situ comparisons would be helpful to further constrain properties of large NAT particles.**

    Simulations for single spectra are shown, which are in agreement with the FSSPs and the situation. We added a reference. But probably more additional information is necessary as the FSSPs only measure at flight altitude and an infrared limb sounder observes a large volume from flight alitude up to a few hundred kilometres away.

27. **L480f Here, clarification is required. Since absolute radiances are shown in Figure 11, I would expect that integrated radiances and their differences can be calculated.**

    As MIPAS-Envisat is a satellite instrument and CRISTA-NF an airborne instrument and both have significantly different viewing geometries, spectral resolution, spectral sampling, field-of-view, etc., the absolute radiance values are not the same, even when the same air masses have been observed. Thus, also the differences are not the same and the method cannot be applied. We rephrased the sentence for clarification.

28. **L484 See comment to L476ff: it has been shown before that a similar signature can be simulated by assuming spherical particles. However, close inspection showed significant discrepancies between simulated and observed spectra in the case of spherical particles.** Please see answer to comment 25.

29. **Fig 11 The channel LRS 5 seems not to be shown (compare L99).**
We extended the figure and now show also the region around 950 cm^{-1} in the way as for the simulations.

30. **Fig 10 An additional panel including a map with the geolocations of the observations would be helpful to visualize the location and size of the sampled region.**
We added this panel.

Technical corrections

1. **L32 denitrifaction -> denitrification**
corrected

2. **L115 KOPRA should be expanded**
done

3. **L450 CALIPSO and CALIOP should be expanded and a reference should be provided (e.g. Pitts et al., 2018)**
done

4. **L458 transfered -> transferred**
done

5. **L478 FSSP should be expanded and a reference should be provided**
done

6. **L500 GLORIA should be expanded and a reference should be provided**
done

7. **L508 Sentence should be revisited. How can new PSC observations be obtained from the presented method? (Possibly, "observations" should be replaced e.g. by "data set")**
In the preprint version the sentence is: "... new data set of PSC observations ..."

8. **Fig 3, Fig 6 Plots may be refined: data points seem to overlap strongly and it is not clear whether significant populations of data points are hidden below other data points. At least, this should be discussed in the text.**
We added information to the text.

9. **Caption Fig 4 spetrum -> spectrum** done

10. **Fig 7b The x-axis might be scaled from e.g. -4 to >8 for a better focus on the relevant region** We changed the scale to -4 – 8. As the gradient exceeds 20 only at one point we avoid to show the full positive range to show more details in the other range.