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Atmospheric Measurement Techniques
Discussions

Interactive comment on "Radiative transfer simulations and observations of infrared spectra in the presence of polar stratospheric clouds: Detection and discrimination of cloud types" *by* Christoph Kalicinsky et al.

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General comments

Based on radiative transfer simulations, the authors present a method to detect and discriminate PSCs in 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≤1.0 μm), medium-sized

NAT (median r=1.5 μm – 4.0 μm) and large NAT (median r≥3.5 μm). 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.

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.

3) The spectral range used by the windows for NAT index-1 to -3 focuses only on the spectral region around the feature at ~820 cmˆ-1. In previous work, it has been shown that, beside details of the spectral feature at ~820 cmˆ-1, the overall spectrum and especially the spectral region towards 960 cmˆ-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 cmˆ-1) to constrain the size classification and define the criteria?

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 tropospheric clouds considered for the discussed flight?)

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 10 μm (see Molleker et al., 2014, Table 1), while the proposed size classification suggests STS and medium-sized NAT

(r=1.5 μm – 4.0 μm) 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.

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).

Specific comments

L15, L492 It should be mentioned that the size classification assumes spherical NAT particles.

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 et al., 2017, Tritscher et al., 2019).

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).

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.

L67, L227 “Very small” should be defined.

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.

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

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

L127ff Information on the along-track sampling would be helpful.

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)).

L163 Did the simulations account for CFC-12? (spectral band centered at ~920 cmˆ-1)

L202, L222 Detailed analysis in the Woiwode et al. (2016) showed that the shape of the feature is sensitive to particle shape.

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 cmˆ-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.

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.

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

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 in polar winters (e.g. Pitts et al., 2018).

L392 The motivation for choosing the discussed flight should be provided.

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.

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.

L422 Why are only spectra with CI < 3.0 shown here? (compare Fig. 3 and L388)

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.

L435,436 See 5): Here, the CRISTA-NF observations are classified as STS or medium sized (median r=1.5-4.0 μm) NAT. However, collocated in situ observations suggest particles with radii exceeding 7.5 and 10 μm (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.

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.

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 abovementioned 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=1 μm), spherical and aspherical particle populations show an almost identical spectrum, while at r=3 μm 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.

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 cmˆ-1 and a significant negative slope and/or an "upward arching" 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 (Wag-

ner 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.

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.

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.

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.

Fig 11 The channel LRS 5 seems not to be shown (compare L99).

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.

Technical corrections

L32 denitrifaction -> denitrification

L115 KOPRA should be expanded

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

L458 transfered -> transferred

L478 FSSP should be expanded and a reference should be provided

L500 GLORIA should be expanded and a reference should be provided

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”)

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.

Caption Fig 4 spetrum -> spectrum

Fig 7b The x-axis might be scaled from e.g. -4 to >8 for a better focus on the relevant region

Literature

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