Reply to the comments of reviewer 2 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

The paper describes methods to detect PSCs and to classify them. The following PSC types are distinguished: NAT (nitric acid trihydtrate), STS (supercooled ternary solution) and ice clouds. For the detection of small NAT particles, a spectral feature at about 820 cm^{-1} has often been used. The authors show, that for larger NAT particles this feature shifts towards smaller wavenumbers and they use this feature to further specify the NAT particles as sNAT (small particles), mNAT (medium particles), and lNAT (large particles). Ice is detected by using the difference between 2 spectral regions (~833 cm^{-1} and ~950 cm^{-1}), the radiance is similar in clear sky conditions but significantly smaller at 950 cm^{-1} when ice is present, a well-known feature which has already been used before. When the cloud is neither NAT nor ice, it is classified as STS. Further a method based on the so-called Cloud Index (CI) is developed to derive the cloud bottom height.
The methods are applied to a comprehensive set of radiative transfer simulations including NAT, ice and STS, all with various concentrations and particle size distributions. Based on the simulations it is demonstrated that the methods work well, in particular sNAT can be distinguished very well from the larger NAT particles mNAT and lNAT. Finally the new methods are also applied to observations of the CRISTA-NF instrument taken during a flight of the RECONCILE campaign
Generally the newly developed methods are described quite well and the figures are appropriate. However, I think that some clarifications are needed, in particular the motivation is not so clear for readers who do not work in the specific field of research (see also my comments below). The scope of the paper fits AMT very well, therefore I recommend publication after taking into account the comments below.
General comments:

1. **The motivation is not clear. It is said in the abstract that there are "uncertainties in the representation of PSCs in model simulations". Which models? Atmospheric chemistry models, climate models? Please provide**

some examples of models which can be improved by better knowledge of PSC properties. Which properties are required by these models? What are the most relevant parameters? Composition, concentration, size, shape ...? The methods presented here do not derive number concentrations, can these also be derived from spectral IR observations? How important is the classification into different size regimes?
Especially, chemistry climate models (CCMs) that are used to asses polar stratospheric ozone loss (e.g. Eyring et al. (2013)) often use rather simple schemes to represent PSCs in the model simulations. Such simplifications may lead to a heterogeneous chemistry dominated by NAT, but it is known that heterogeneous chemistry on STS and cold binary aerosol particles probably dominates the chlorine activation (e.g. Solomon (1999), Drdla and Müller (2012), Kirner et al. (2015)). Additionally, no comprehensive microphysical models are typically used to describe evolution of PSCs over the winter. Mesoscale temperature variations that are known to play an important role for the formation of PSCs (Carslaw et al. (1998), Dörnbrack et al. (2002), Engel et al. (2013), Hoffmann et al. (2017)) are also missing in current state of the art CCMs (Orr et al. (2015)).
Assumptions on the occurence of different PSC types typically have only limited impact on many aspects of ozone loss, as, for example, liquid PSC particles are sufficient to simulate nearly all ozone loss (Wohltmann et al. (2013), Kirner et al. (2015), Solomon et al. (2015)). However, there are also situations where the PSC type is crucial. For example, which PSC type is present at top of ozone loss region is important (Kirner et al. (2015)) and during the initial activation in PSCs covering only a small part of the vortex the type plays also an important role (Wegner et al. (2012)).
Furthermore, the heterogenous reaction rates on PSCs strongly depend on temperature but also on the PSC type (e.g. Drdla and Müller (2012), Wegner et al. (2012)). Here, especially for NAT the reaction rates show rather large uncertainties (Carslaw et al. (1997), Wegner et al. (2012)), which highlights the importance of observing the PSC type.
In summary, information on the compostion of the PSCs is very important, but measurements are limited. Typical PSC measurement techniques are in-situ particle measurements and lidar observations (e.g. Molleker et al. (2014), Achtert and Tesche (2014), Pitts et al. (2018)). But beside these measurement techniques an infrared limb emission sounder builds a good basis for such kind of studies.
We added these information to the introduction.

2. **Cloud optical thickness is a useful parameter to describe a cloud, the term is sometimes used in the paper but no number is given. What is the optical thickness range of the clouds considered here (in the RT simulations and what can be observed using the limb observations)? I suppose that all clouds are optically very thin, what is the upper limit of cloud optical thickness that can be analysed with the presented methods?**
We added the extinction ranges to Tab. 3 for each individual PSC type. They range from 1.0e^{-5} – 1.0e^{-2} km^{-1}, whereby the largest extinction is only achieved by ice. For the optical thickness (= extinction * vertical thickness) the range is then 0.5e^{-5} to 8.0e^{-2} for ice, from 1.05e^{-5} to 3.3e^{-2} for STS and from 1.7e^{-5} to 1.76e^{-2} for NAT.
As the clouds are typically detected using the CI and a threshold value (here 3.0), the

upper limit with respect to the optical thickness cannot be stated here. As the CI depends on many different factors (e.g. cloud type, particle radius, altitude of the cloud etc.), the CI values for the same extinction and optical thickness can be different (see General comments Reviwer 3). Thus, the upper limit also depends on these factors influencing the CI and cannot be stated in general.

3. **The radiative transfer simulations are based on a single scattering approach which is not described in detail here. This also means that it is only valid for very thin clouds, for which multiple scattering can be neglected (see e.g. Höpfner and Emde 2005). Please provide the optical thickness range used for the radiative transfer simulations to justify the neglect of multiple scattering.**
   We compared our results with the findings by Höpfner and Emde (2005) and estimated the maximum uncertainties that can occur. In the case of STS the SSA of 0.24 also fits to our simulations. The scenarios defined in Höpfner and Emde (2005) that fit to our simulations are typically 1 and 2, maybe for a small part of the simulations it is scenario 3. This leads to uncertainties typically $\leq 1\%$ (4.5% for some simulations). In the case of NAT the scenarios are 1 and 2. The SSA lies between the two SSA simulated by Höpfner and Emde (2005) of 0.24 and 0.84. Here we took a mean SSA of 0.54 and a mean uncertainty of the both analysed SSAs. Then for NAT the uncertainty is $\leq 4\%$. For ice the scenarios are mainly 1 and 2 and for a small portion of the simulations (those with the largest volume density) also scenario 3 fits. The SSA is comparable to NAT and we used 0.54. This leads to uncertainties $\leq 4\%$ ($\leq 20\%$ in case of sceanrio 3). In total for almost all simulations the uncertainties are $\leq 4\%$ and only a few simulations have larger uncertainties. But with respect to the computational effort these small uncertainties do not justify the use of multiple scattering.
   Furthermore, in our analysis we typically use radiance ratios. As the single scattering approach leads to an underestimation of the radiance that is often similar in many spectral regions, the uncertainties of the ratios are much smaller than the uncertainty of the radiances themselves. E.g. an underestimation of the radiance by 10% in all spectral regions would lead to the same ratios.
   We added information to the Sect. 2.2.

4. **Presuambely, the JURASSIC model does not account for horizontal inhomogeneities. Please discuss the validity of this approach for PSCs, i.e. how large is the horizontal extend of the PSCs typically compared to the line of sight through the PSCs?**
   Our simulation setup does not account for horizontal inhomogeneities of the PSC. The horizontal extent of the line of sight of the instrument inside the PSC can reach up to several hundred kilometres. In case of synoptic scale PSCs horizontal homogeneity is a good approximation. Other events, such as e.g. mountain wave ice, can lead to PSCs with a smaller horizontal extent. But with respect to the large amount of PSCs simulated in this study we started with the most simple approach regarding the horizontal homogeneity of the PSCs.
   We added some information to Sect. 2.2.

5. **For the reader it is rather difficult to remember the definition of all indices used in the paper. I think it would be helpful to include a figure showing**

the spectral regions used and mark the spectral windows that are used to calculate the individual indices. Also a table including the definitions of NAT-indices 1,2,3 and the CI index could be useful.
We inserted the different regions into Fig. 1 and numbered the micro windows (MW) from 1 to 7. We additionally added a new table for all indices, where the ratios and BTD that are used can be seen.

6. **I miss some discussion about the uncertainties. For example on p.8 you write: "Nearly all simulations with NAT particles $< 3\mu$m lie above the region of the simulations for STS and ice clouds, which is marked by the solid black line. Thus, NAT particles within this size range can be detected and discriminated using NAT index-1." For the modelled spectra this is correct, but also for real observations? Measurements always include uncertainty, how accurate measurements are required so that NAT is clearly separated from the ice clouds?**
Because of the strong radiance enhancement caused by the clouds, the dominating uncertainty is the relative uncertainty (noise uncertainty) of the measurements, whereas a systematic uncertainty (e.g. an offset) plays a minor role. The relative uncertainty for the radiances typically observed during atmospheric measurements is about 1–2% for CRISTA-NF (Schröder et al. (2009)). Because of the calculation of ratios, the relative error can add up but is still only a few percent. In the worst case (one MW plus 1-2% and the other MW minus 1-2%) the maximum uncertainty is about 2-4%. For the analysis of the PSC observations this has a different effect depending on the CI and the NAT-index. For smaller values of these quantities the resulting absolute uncertainties of the quantities determined from the relative uncertanties of maximum 4% are smaller than for larger values, e.g. a CI of 2 has a maximum uncertainty of ± 0.08 and a CI of 5 an uncertainty of ± 0.2. Thus, the methods work better for smaller CI values. This suggests to use a threshold value for the CI and to only analyse observations with a CI below this threshold. This suggestion we consider supported by our finding that the separation between the different size bins was better for a CI below 3 (Fig. 3 and Fig. 4). So we keep this value. Furthermore, the same spectral windows are used for CI and NAT index-1/2. In this case some of the uncertainty will cancel, as the data points will shift similar to the correlation regions/lines, i.e. a smaller CI (because of a smaller randiance in the CO_2-window) will be accompanied by a larger NAT index and vice versa. Lastly, the scatter plots for the observations (new Fig. 9), especially for flight 3 and 5, show that a large number of observations exhibit deviations from the separation line larger than a few percent and a clear separation between ice (flight 1) and NAT is visible in Fig. 9 a) and b). Furthermore, the measurements show a compact correlation, which we consider an indications for little noise, whereas we would expect more spread for large noise errors.
Information on the uncertainties are added to the text.

Specific comments:

1. **The title is a bit misleading. The main focus of the paper are the methods to classify and detect PSCs. I thought from reading the title starting with "Radiative transfer" that the paper was more about radiative transfer methods etc. For the RT simulations a well-known model JURASSIC is applied**

but the methodology is not described in this paper. Also the observational methods are not described here. I think that the title should be something like e.g. "A new method to detect and classify polar stratospheric clouds"
We changed the title to:
A new method to detect and classify polar stratospheric NAT clouds derived from radiative transfer simulations and its first application to airborne IR limb emission observations

2. **Abstract: p1, l7 "... showed a spectral peak at about 816 cm-1 . This peak is shifted compared to the peak at about 820 cm-1, which is known to be caused by small NAT particles. " -> A bit more information about this peak would be helpful. What is the physical process responsible for the peak. Which physical processes could produce a shift of a spectral peak ...**
The peak at 820^{-1} is mainly caused by emission of radiation. The transformation then is caused by the increasing contribution of scattering to the total extinction. We briefly added this to the abstract.

3. **p1, l16: "gradient of the CI" -> which gradient is meant here? -> "vertical gradient"**
yes, we corrected this

4. **p3, l31-33: These 3 sentences should be shifted to Section 2.2**
These 3 sentences are only a short summary was is comming in the long Sect. 2 in total. The details are then given in the subsections and Sect. 2.2 describes the radiative transfer code JURASSIC. We find that these sentences do not fit to Sect. 2.2. Thus, we stay with the old text.

5. **p4. l19: Title of section should include the term "radiative transfer simulations"**
we changed the title to "Radiative transfer simulation code JURASSIC"

6. **p4 l32: "The optical properties of the particles, extinction coefficient, scattering coefficient, and phase function, required for the radiative transfer simulations ..." -> "The optical properties of the particles (extinction coefficient, scattering coefficient, and phase function) required for the radiative transfer simulations ...", include brackets here because extinction coefficient, scattering coefficient, and phase function are the optical properties**
done

7. **p7 l20: What is the "scattering radius" of a particle, this term has not been defined**
The scattering behaviour of a PSD depends on its median radius μ and distribution width σ and the wavelength. In many cases the effective radius of a PSD is a sufficiently good approximation to describe the scattering behaviour with a single parameter. However, when particle size and wavelength are approximately the same size, this is not a good approximation as the scattering behaviour for two PSDs with same r_{eff} but different μ and σ can be different. Here, the scattering radius r_{sca} is a better single

parameter to describe the scattering behavior:

$$r_{eff} = \frac{\int_{r_0}^{r_1} \pi r^3 n(r) \mathrm{d}r}{\int_{r_0}^{r_1} \pi r^2 n(r) \mathrm{d}r} \tag{1}$$

$$r_{sca} = \frac{\int_{r_0}^{r_1} \pi r^3 n(r) Q_{sca} \mathrm{d}r}{\int_{r_0}^{r_1} \pi r^2 n(r) Q_{sca} \mathrm{d}r}, \tag{2}$$

where Q_{sca} is the scattering efficiency depending on radius r, wavelength and complex refractive index e.g. see Hansen & Travis (1974). Since we assumed only one σ in our simulations the PSD median radius is sufficient to unambiguously characterise the scattering behaviour in our study.

We revised the sentence:
“ The appearance of the spectral feature that is observed in infrared limb spectra in the presence of polar stratospheric clouds consisting of NAT particles in our study is unambiguously characterized by the median radius of the particle size distribution, as we kept the distribution width σ constant.”

8. **p7 l31: "different contributions of extinction and scattering" -> extinction is the sum of absorption and scattering, therefore I think that you mean "absorption and scattering"**
yes, we corrected this

9. **Fig.1 and 5: "spectra have been scaled such that the radiance equals 1" -> scaling factor is not clear, is it the average radiance over the plotted spectral range?**
The scaling factor is 1/(mean radiance in window 832-834m^{-1}). We added this.

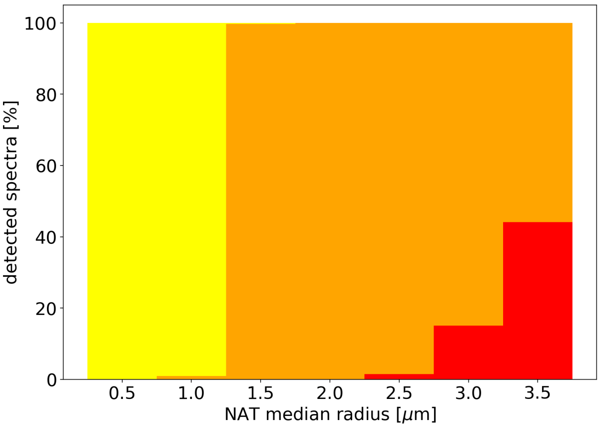
10. **Fig.2: For the interpretation of the RT simulations, it would help to include here also the refractive indices of ice and STS. Further I think that it would be very helpful to show extinction and absorption coefficients, which are probably calculated using Mie theory for the individual PSC types and for some particle sizes to see, that for larger particles the scattering coefficient dominates.**
We added the refractive indices for STS and ice. Furthermore, we show two more plots now, the extinction and the SSA for NAT with different median radii. These plots illustrate the behaviour of the radiance spectra with increasing median radius. We rephrased the text in Sect. 3.1 accordingly.

11. **p9, l4: "results for PSCs with larger NAT particles (up to 4μm) also lie above the simulations for STS and ice (black separation line)": In Fig.3 it is not well visible, which radii lie above the separation line. May be discrete colours could be used for each of the simulated radii?**
We used new discrete colours for the Fig. 3. Additionally, we also changed the colours in Fig. 1, Fig. 2, and Fig. 6 to use always the same colours for the NAT, STS, and ice simulations.

12. **p9, l23: "detected NAT spectra" -> NAT is detected, not the "spectra"**
we rephrased the sentence

13. **p10, l23: "When the NAT detection and classification procedure (described in the previous subsection) is applied to the simulation results for the mixed clouds, the good discrimination between the small and medium size particles remains." -> this is not shown, why?**

    The plot is limited to the range 0.5 to 3.5μm as only a subset of the NAT simulations is combined with STS. Thus, we decided to give the most important numbers regarding the detection capacity in the text and show the plot here in the reply.

14. **p13, l3-9: Here you discuss about "cloud optical thickness" (thick and thin) without providing any values -> as mentioned above, please quantify the cloud optical thickness**

    The terms thin and thick in this context are relative terms. Unfortunately, a clear relationship between the optical thickness and the CI does not exist (see General comments Reviewer 3). As all different clouds (different type, radius, altiude etc.) enter the analysis here the optical thicknesses can be different for the same CI values.

    Nonetheless, we rephrased the text to clarify the meaning. There are two cases where a bottom altitude cannot be detected. In some cases the observations run into saturation and the CI values below the cloud bottom altitude stay very low. Additionally, in some other situations the CI values below the bottom altitude only show a linear increase and not the larger change directly below the bottom altitude. In both cases the approach fails. A way to sort out such possibly affected observations is the use of a threshold value. For our simulations a CI minimum of 1.25 turned out to be sufficient. Such low values of CI only occur for a part of the ice clouds simulated here (typically that with the largest volume densities and for a few of the NAT and STS clouds with large HNO_3 VMRs ($\geq$ 11 ppbv) or volume densities ($\geq 5\ \mu\ m^3/cm^3$) combined with a large vertical thickness ($\geq$ 4 km).

15. **p15, l7: "... where much more NAT was observed by CALIOP (the difference in the Southern hemisphere is much smaller)"-> how much more NAT was observed by CALIOP, how much smaller is the difference in the Southern hemisphere?**

    The agreement for NAT in the Southern hemisphere is 73%, whereas there is only an agreement of 18% in Northern hemisphere. We added these numbers to the text.

16. **p16 l20: "This method can surely be transferred to other cloud observations**

such as cirrus clouds and aerosol layers and to other airborne instruments measuring in the same wavelength region like e.g. GLORIA." -> Which clouds and aerosols could be observed? Probably only very thin clouds? Up to which cloud optical thickness can the method be applied?
As said before, our method rely on the CI and there is no clear relationship between the CI and the optical thickness (see General comments Reviewer 3). Thus, a clear statement cannot be made as the upper limit depends on many factors.