We thank the reviewer for providing constructive comments. Please find below our answers, along with some summary descriptions of the modifications we made in the revised manuscript we are submitting. For clarity, we also include, **in bold**, the original reviewer's comments.

Summary

This paper presents a sophisticated scheme for modeling ice cloud optical properties by incorporating mixed ice crystal habits, leveraging the upcoming FORUM experiment data. It tests the retrieval scheme on simulated measurements, demonstrating promising performance and identifying the main error components affecting cloud parameter retrievals.

Strengths

• Innovative Approach:

Develops a new methodology for retrieving ice crystal habit mixtures from spectral radiance measurements, filling a significant gap in atmospheric science.

• <u>Utilization of FORUM Data</u>: The study is forward-looking, leveraging future FORUM experiment measurements to improve ice cloud parameterization, which is crucial for climate modeling.

We thank the reviewer for the comments and for emphasizing the strengths of the technique we presented in our manuscript.

Weaknesses

• Ill-posed problem:

The concern regarding the manuscript's handling of the ill-posed problem, particularly the issue of solution uniqueness, is a critical aspect that warrants further clarification and enhancement in the manuscript. The ill-posed nature of the problem stems from the inversion process involved in retrieving ice crystal habit fractions from spectral radiance measurements, which, in some scenarios, is shown to be particularly challenging due to the possibility of multiple solutions yielding almost the same minimum of the cost function. I suggest to the authors to expand this point. This will enhance the manuscript strength, providing a more comprehensive understanding of the illposed nature of the retrieval problem.

At the first run of our test retrievals, we found that about 12% of the cases were missing the convergence to the absolute minimum of the cost function (CF). This issue was attributed to the ill-posed nature of our inversion problem. In general, the relatively weak sensitivity of the measurements to the retrieval parameters

decreases the curvature (i.e. makes "flat") the hyper-surface of the CF of which we want to find the absolute minimum. Moreover, the measurement noise and the forward model errors introduce a sort of "roughness" in the CF hyper-surface. Finally, if some parameters included in the state vector are correlated to each other, narrow "canyons" may also characterize the hyper-surface of the CF (see Transtrum et al. 2011 and Ridolfi and Sgheri, 2013). All-together, these elements make the inversion problem ill-posed, and the search of the absolute minimum of the CF becomes a challenging task. The Gauss-Newton method modified with the LM damping, being based on the CF gradient may get trapped on a secondary, local minimum, as actually happened in our first retrieval attempts. A solution could have been to use a stochastic method in place of the LM to find the CF minimum. For example, in Ridolfi and Sheri (2009), the simulated annealing method was used to find the absolute minimum of a CF specifically designed to find the optimal strength of the height-dependent Tikhonov regularization applied to the retrieval of vertical profiles from limb sounding spectral measurements. The simulated annealing, however, as other stochastic minimization methods, requires thousands of evaluations of the CF. In our case, the evaluation of the CF implies the evaluation of the forward model, thus a computationally heavy operation. This feature clearly makes stochastic methods inadequate for our application.

- We managed to overcome almost completely this convergence issue by repeating a few times the retrieval starting from slightly different initial guesses of the state vector and by selecting, a posteriori, the solution corresponding to the smallest value of the cost function. With this strategy, only 4 out of the 375 retrievals presented actually miss the absolute minimum of the CF.
- It is worth to note that in the simulated retrievals, the cases with a large convergence error are easily detected because in these cases a difference much larger than the error bar exists between the retrieved and the true parameter values. In real data analysis the problem may be harder to identify. A strategy could be to compare the retrieved parameter values and the achieved CF minimum with the respective accumulated statistics, and to treat the outliers as "suspicious" cases. In these cases, restarting the retrieval from different a priori parameter estimates could be beneficial, as it proved to be in the test retrievals presented in the paper. Also a visual inspection of the residuals of the fit for some selected suspicious cases could help to diagnose the problem and to find a workaround.

We included these comments / explanations in Sect. 3.2 of the revised paper.

• Limited Validation:

The methodology is tested only on simulated data, lacking validation against actual satellite measurements, which raises questions about its real-world applicability.

- Yes, this is a new method that certainly needs to be validated against real measurements, however, we consider such an extensive validation beyond the scope of the current paper. In the near future, we plan to apply the developed retrieval scheme to the large dataset of ground based spectral radiance measurements collected in Antarctica by the Radiation Explored in the Far-InfraRed - Prototype for Applications and Development (REFIR-PAD), a Fourier spectroradiometer deployed by our institute since December 2011 (Palchetti et al., 2015). These measurements are complemented by co-located backscattering/depolarization lidar measurements permitting to estimate the cloud geometrical extension, and by the statistics of ice crystal shapes determined from the measurements of an ice camera deployed on the same site. Potentially, the synergy of these measurements could allow to validate the inversion method presented in this work on the basis of real data and, at the same time, to build a climatology of Antarctic ice cloud crystal habits, corroborated by local in situ measurements.
- In the manuscript, we state the need to validate the proposed method against real measurements both in the introduction and at the end of the conclusion section. In the revised version of the manuscript, we further tried to clarify this point.

• <u>Complexity and Accessibility:</u> The complex methodology and reliance on specific satellite data may limit its accessibility and applicability by the broader research community.

The complexity of the methodology is mainly linked the full-physics model embedded in the proposed method. Simpler methods are easier to use but the information extracted from the measurements is necessarily less exhaustive. In conclusion, we agree that the algorithm proposed may be implemented and exploited only by highly specialized scientists of the field. On the other hand, the method may be applied to the measurements of the forthcoming FORUM and PREFIRE satellite missions, with the potentiality to derive accurate statistics of cloud properties that are extremely relevant for climate studies. In principle, the method could also be applied to the current IASI measurements. However, since IASI measurements are limited to the mid-infrared spectral region, it is not certain whether these measurements contain sufficient information to disentangle the contributions of the various ice crystal shapes to the upwelling spectrum. In the conclusions of the revised manuscript, we added a comment on this possible application of the proposed method.

• <u>Clarify Methodological Assumptions and Limitations:</u> The manuscript would benefit from a more detailed discussion of the assumptions underlying the SACR code and the optimal estimation approach used. Addressing the potential limitations these assumptions may pose to the generalizability of the findings will strengthen the manuscript. For instance, how might different atmospheric conditions or cloud compositions affect the retrieval accuracy?

- In the paper, we show that for the mid-latitude and tropical scenarios the algorithm works very well assuming the noise level expected for FORUM measurements. The performance gets worse in polar scenarios, because in these cases clouds are often very close to the ground (on the Antarctic Plateau the ice clouds may be placed even a few hundred meters above the ground) and have a temperature close to that of the ground. In these conditions the cloud emission becomes indistinguishable from that of the ground, thus the inversion becomes strongly ill-conditioned and the probability for the minimization procedure to get trapped into a secondary minimum of the cost function is larger. In these cases the retrieved cloud parameters differ from their true value beyond their error bar. In the revised version of the paper, we discuss how these occurrences could be detected and mitigated in real data analysis (see Sect. 3.2 of the revised paper).
- <u>Strengthen the Literature Review</u>: A more thorough review of the current state of research in ice cloud characterization and the retrieval of cloud properties from spectral radiance measurements could provide a stronger foundation for the study. Highlighting the novelty of the approach in the context of existing methodologies will help to underscore the contribution of this work to the field.

We added the following references in the introduction of the manuscript:

[1] Lolli, S., Campbell, J. R., Lewis, J. R., Gu, Y., and Welton, E. J., Fu-Liou-Gu and Corti-Peter model performance evaluation for radiative retrievals from cirrus clouds. *Atmospheric Chemistry and Physics*, 17(11), 7025-7034, (2017).

[2] Campbell, James R., Erica K. Dolinar, Simone Lolli, Gilberto J. Fochesatto, Yu Gu, Jasper R. Lewis, Jared W. Marquis, Theodore M. McHardy, David R. Ryglicki, and Ellsworth J. Welton. "Cirrus cloud top-of-the-atmosphere net daytime forcing in the Alaskan subarctic from ground-based MPLNET monitoring."Journal of Applied Meteorology and Climatology 60, no. 1 (2021): 51-63.

[3] Lewis, J. R., Campbell, J. R., Stewart, S. A., Tan, I., Welton, E. J., and Lolli, S. (2020). Determining cloud thermodynamic phase from the polarized Micro Pulse Lidar. *Atmospheric Measurement Techniques*, *13*(12), 6901-6913.

Minor Comment

• Graphical Representations: Some figures are dense and may be simplified for better clarity and comprehension.

Recommendation

Given its innovative approach and potential impact on climate modeling, I recommend this paper for acceptance with minor revisions. Addressing the validation with actual satellite data and simplifying complex explanations and figures would significantly enhance the paper's value and readability.

As mentioned in our answer to the above reviewer's comment titled "Limited validation", in the near future we will extend the validation of our proposed method to cover also the analysis of real measurements. This operation, on its own, is a huge task that we believe is beyond the scope of this paper that, in our view, should be limited to the description of the theoretical basis of the method and on testing its self-consistency and performance on the basis of realistic, synthetic measurements.

The paper we are re-submitting was revised according to the constructive reviewer's comments supplied and, we believe, now should be much easier to read than the original version.

References

Ridolfi, M. and Sgheri, L.: A self-adapting and altitude-dependent regularization method for atmospheric profile retrievals, Atmos. Chem. Phys., 9, 1883–1897, https://doi.org/10.5194/acp-9-1883-2009, 2009.

M. Ridolfi and L. Sgheri, "On the choice of retrieval variables in the inversion of remotely sensed atmospheric measurements," Opt. Express **21**, 11465-11474 (2013).

M. K. Transtrum, B. B. Machta, and J. P. Sethna, "Geometry of nonlinear least squares with applications to sloppy models and optimization," Phys. Rev. E 83, 036701 (2011).