Reply to referee #2: Interactive comment on "Retrieving ice nucleating particle concentration and ice multiplication factors using active remote sensing validated by in situ observations" by Jörg Wieder et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-67-RC1, 2022

Reviewer comments are reproduced in **bold** and author responses in *italic*; extracts from the original manuscript are presented in *red italic*, and from the revised manuscript in *blue italic*.

Review for Wieder et al., "Retrieving ice nucleating particle concentration and ice multiplication factors using active remote sensing validated by in situ observations", submitted to Atmospheric Chemistry & Physics

This manuscript presents a method to retrieve ice-nucleating particle (INP) concentrations using a polarization Raman lidar and a Ka-band cloud radar as well as ice multiplication factors due to secondary ice production in orographic mixed-phase clouds. The retrievals were compared against in situ observations derived from a tethered balloon system at two locations: the WFJ and WOP sites at different altitudes during the RACELETS field campaign in the Swiss Alps.
Retrievals of INP concentrations and ice multiplication are both extremely poorly constrained and valuable for better constraining cloud properties. The study is thus well-motivated and using tethered balloons for ice crystal number concentration is advantageous compared to aircraft in situ probe measurements because they don't suffer from ice crystal shattering effects. However, details describing the methodology and a clear disclosure of assumptions and quantification of limitations is lacking. Specific comments follow.

We would like to thank the Referee 2 for reviewing our manuscript. We are pleased with the positive reception and grateful for the helpful comments which improved our manuscript and are answered individually hereafter.

The method is not described in sufficient detail. This is especially important given the large number of assumptions that need to be made in the retrievals. For example, the method to retrieve the INP number concentration is appears to use the various INP concentrations, but not a single equation for any of the parameterizations appears in the manuscript. The same goes for the ICNC retrieved by the radar under the assumption of a particular ice crystal size distribution which was not described.

We thank the reviewer for pointing out the need for more details about the used methods. We extended the methods section accordingly, especially by providing more details on the lidar retrieval of aerosol properties (Section 2.2), the retrieval of ice crystal properties from both remote sensing (Section 2.3) and the in situ instrument (Section 2.4), and explicit equations of all used INP parameterizations (Section 2.5). Due to the large number of changes please refer to the updated track changes version of the manuscript.

Error quantification is almost nonexistent in this work.

We thank the reviewer for pointing out the need for more error quantification as we have mostly put an emphasis on the discussion of the induced methodological uncertainties in our manuscript. We provided the errors of all used quantities in the method section (Sections 2.1, 2.2, and 2.3) and indicated them at meaningful positions in different plots, see Figures 2, 3, 4, 5, 6, 7, and 8 in the revised manuscript.

Why does the lidar almost always overestimate the in situ observations at the WFJ site? I couldn't find an explanation for this.

We thank the reviewer for pointing out the need for clarification. The remote sensing measurements in the very clean air above the Alps are challenging. The previously used conversion factors from the lidar to aerosol concentrations (see Equations 2 and 3 in the revised manuscript) were partially responsible for an overestimation of the aerosol concentrations. We derived new conversion factors from the dataset collected during

our campaign which reduced the retrieved concentrations by up to 20 %. Furthermore, given the very low extinction coefficients in the clean air over the Alps, the assumed linear relationship by Mamouri and Ansmann (2016) in Equations 2 and 3 may not hold anymore. An overestimation in the remote sensing observations has also previously been reported by Haarig et al. (2019) using the similar instrument in a Saharan dust layer at Barbados. We extended the argumentation in lines 285-296 (revised manuscript) to:

At relatively low aerosol concentrations and in the presence of continental aerosol, a plateauing of the lidar-retrieved aerosol concentrations was observed. The clear atmosphere over the Alps with very low values of the extinction coefficient (< 10 Mm^{-1}) could be responsible for deviations from the assumed linear relationship of extinction to n_{250} and s, respectively, (Mamouri and Ansmann, 2016). The larger diurnal variability between the in situ observation and lidar retrieval at WOP (high valley site) compared to WFJ (mountaintop site) can be explained by the diurnal changes of aerosol concentration near the ground (in situ observations) not affecting the air masses on the lidar retrieval height (height difference approx. 400 m, see Figure 1b). This difference in height and therefore air mass commonly limits a quantitative conclusion between ground-based in situ observations and remote sensing instruments as the well-mixed boundary layer could at times not extend up to the lowest retrieval height. For the retrieval of s (Figures 2c and 2d) the aforementioned observations hold equally true, which is not surprising as the surface area relates to the square of particle radius. However, comparing the retrieval accuracy at WFJ (Figures 2a and 2c), a stronger bias of the lidar retrieved surface area concentrations is apparent which has been also previously reported by Haarig et al. (2019), based on similar observations carried out at Barbados.

For the non-expert in in situ measurements, why does the ambient air need to be heated to 46C in the inlet?

We thank the reviewer pointing out the need for more explanation. We extended the discussion by lines 137-142 (revised manuscript) as follows:

At both sites, ambient air was sampled through a 46°C-heated inlet. The heating was a preventive measure to avoid icing of the outside inlet parts, to evaporate activated cloud droplets, and to sublimate ice crystals. The evaporation of volatile compounds of the aerosol cannot fully be excluded. However, the effect is expected to be minor given the high flow rate through the inlet (300 L min^{-1}), such that the temperature of sampled air was likely below 46 °C. Furthermore, the degradation of relevant INPs (mostly biological) should only occur at temperatures above 46 °C (Kanji et al., 2017; Huang et al., 2021) and is hence regarded as unlikely (see Wieder et al., 2022b, for further details).

References

Haarig et al. (2019), Huang et al. (2021), Kanji et al. (2017), and Mamouri and Ansmann (2016) as in manuscript.

Wieder, J., Mignani, C., Schär, M., Roth, L., Sprenger, M., Henneberger, J., Lohmann, U., Brunner, C., and Kanji, Z. A.: Unveiling atmospheric transport and mixing mechanisms of ice-nucleating particles over the Alps, Atmospheric Chemistry and Physics, 22, 3111–3130, https://doi.org/10.5194/acp-22-3111-2022, https://acp.copernicus.org/articles/22/3111/2022/, 2022b.