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Supporting information for

On the links between ice nucleation, cloud phase, and climate sensitivity in CESM2

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Contents of this file:

Text S1
Figures S1 to S10
Supplementary References

Text S1. Individual mechanisms for INP influences on cloud feedback strength

Here we present our interpretations for each individual influence of mixed-phase microphysics on cloud feedback strength not described in the main text. While we do not isolate with certainty the cause of each microphysics-feedback connection, or evaluate the extent to which our findings are dependent on our model and methodological setup, we present our interpretations here so these may be further explored by other researchers.

Our interpretations are based on a radiative kernel feedback decomposition described in the main text. We include in this supplement detailed visualizations of the decomposed feedbacks, including by mechanism and pressure level (Fig. S5) and both of these factors plus latitude (Fig. S6). We additionally present the raw ISCCP simulator model output (Figs. S7-10) used in conjunction with the radiative kernels (depicted in simplified form in Fig. S3) to carry out the feedback decomposition.

S1a. Cloud amount feedbacks in low and mid-level extratropical clouds in Group A

Adding INPs is found to enhance a positive cloud amount feedback in low clouds yet reduce this feedback in mid-level clouds (see blue bars on the left sides of the 2nd and 3rd panels of Fig. S5). The cloud amount feedback at both levels may be a poleward extension of the established decrease in tropical cloudiness as warming occurs (Bony & Dufresne, 2005). Note that the low and mid-level cloud amount feedback sensitivities in our simulations cancel, and hence do not in sum have a large effect on total cloud feedback. However, they are notable in that they cause cloud feedback differences to be largely attributable to mid-level clouds rather than low (see Fig. 2b). The counteracting amount feedback sensitivities we find may at least partly reflect artifacts of the decomposition method, rather than real behavior. Clouds tend to rise as the atmosphere warms (Hartmann & Larson, 2002), which could potentially displace some clouds from our low cloud to mid-level cloud definitions, creating spurious feedback sensitivities.

That said, our Group A experiments show INPs to alter the present-day cloud occurrence oppositely in low and mid-level clouds. This could influence the potential for existing low and mid-level amount feedbacks to operate, possibly explaining the apparent sensitivities to INPs as real differences among experiments. Looking at the ISCCP cloud type histogram differences in Fig. S7, it is apparent that adding INPs increases the proportion of optically thin low clouds while decreasing the proportion of optically thick mid-level clouds. As both low and mid-level cloud amount feedbacks are positive with no INPs (see blue bars in Fig. S5), increasing (decreasing) the base state cloudiness at a level would be expected to result in a stronger (weaker) positive feedback, which matches the direction of the sensitivities estimated by the refined kernel decomposition.

Why would addition of INPs increase low cloud occurrence while decreasing mid-level cloud occurrence? Impacts of INPs are complex and may result in more or less stable clouds depending on conditions. The presence of INPs can reduce cloud lifetime by enabling the WBF process to deplete cloud liquid, which can grow ice crystals sufficiently heavy to fall and deplete clouds. Contrarily, for a cloud that might otherwise deplete quickly as it transitions from liquid to ice, a large supply of INPs can prolong cloud lifetime by acting as a sufficient number of surfaces for the cloud's liquid mass to be distributed on, preventing ice crystals from falling quickly. The opposite balances across levels possibly result from the cloud depletion effect having less influence in generally shallow low clouds than in mid-level clouds that tend to be more vertically developed, though further effort will be needed to cement the reason. Overall, impacts of model microphysics setup on cloud spatial distribution appear to influence cloud amount feedbacks. Since INPs are shown to affect this distribution, this may be an underappreciated connection between INPs and cloud feedback.

S1b. Cloud amount feedbacks in low and mid-level extratropical clouds in Group B

The above-described low and mid-level cloud amount feedback sensitivities to INPs are apparent in Group B simulations (see blue bars on the right sides of the 2nd and 3rd panels of Fig. S5). This suggests that these impacts of INPs are not solely dependent on INPs' influence on base state global mean cloud phase. However, impacts of INPs on cloud amount feedbacks are weaker in Group B simulations than Group A simulations, suggesting the stronger WBF process in Group A simulations enhances the impact of INPs on base state cloud amount and its associated feedback (see discussion of base state cloud amount and its influence on cloud amount feedback in Text S1a).

S1c. Cloud optical depth feedbacks in low and mid-level extratropical clouds in Group B

Group B experiments with more INPs have stronger cloud phase feedback (see red bars on the right side of the 2nd row of Fig. S5, with this also evident in the ISCCP simulator model output shown in Fig. 10). This is in spite of similar present-day global cloud phase among these experiments (see Fig. S1a). We attribute these cloud phase feedback differences to the WBF adjustment that we enacted in Group B to negate base state global cloud phase differences (see Methods), rather than to the ice nucleation differences themselves. Group B simulations with the most INPs (*Hoose-cap2 (B)* and *Meyers (B)*) have a high potential for INP-formed ice clouds while also having a reduced chance of having their lifetimes shortened through WBF-initiated precipitation. This combination may be the cause of these simulations' high ice water path values (see Table 1). High base state ice water path appears responsible for *Hoose-cap2 (B)* and *Meyers (B)* having the most negative low cloud optical depth feedback among Group B experiments. As with low SLF, high ice water path may enable heightened potential for cloud deglaciation and associated brightening as warming occurs. As model microphysical differences affect ice water path, this can be interpreted as a connection between mixed-phase microphysics and feedback strength. Ice water path is poorly constrained in observations (see range in Table 1), and our results suggest a refined constraint and its incorporation in models may be necessary to reduce cloud phase feedback uncertainty.

S1d. Cloud optical depth feedbacks in high tropical clouds in Group B

Our Group B experiments demonstrate that high cloud optical depth feedback is sensitive to mixed-phase clouds microphysics, which has not previously been examined. In experiments with abundant INPs, particularly *Meyers (B)* and *Hoose-cap2 (B)*, high cloud optical depth feedback is less negative than in other Group B experiments (see red bars on the right side of Fig. S5, bottom panel). This could potentially indicate that cloud phase feedback extends to high clouds in addition to low and mid-level (discussed in Text S1c).

That high cloud optical depth feedback differences are less apparent among Group A experiments suggests this is a consequence of the WBF adjustment we perform only in Group B to maintain similar base state global mean SLF among experiments. Note that adding INPs while weakening WBF results in higher present-day SLF near the equator when global mean SLF is kept constant (shown in Table 1 and Fig. S1b). If these clouds undergo warming with climate change, rather than fully rise with the isotherms (Hartmann & Larson, 2002), the higher base state tropical SLF could enhance the number of high tropical clouds that deglaciate and brighten. Equivalent to how optically thicker low clouds cause increased net cooling, optically thicker high clouds cause net warming (opposite outcome since high clouds tend to be net warming in their base state, as shown in Fig. S3). This is compatible with the sign of the high cloud optical depth feedback difference comparing *Meyers (B)* and *Hoose-cap2 (B)* to the other Group B experiments. We hence interpret the sensitivity of high cloud optical depth feedback to mixed-phase microphysics as reflecting different realizations of SLF's global spatial distribution.

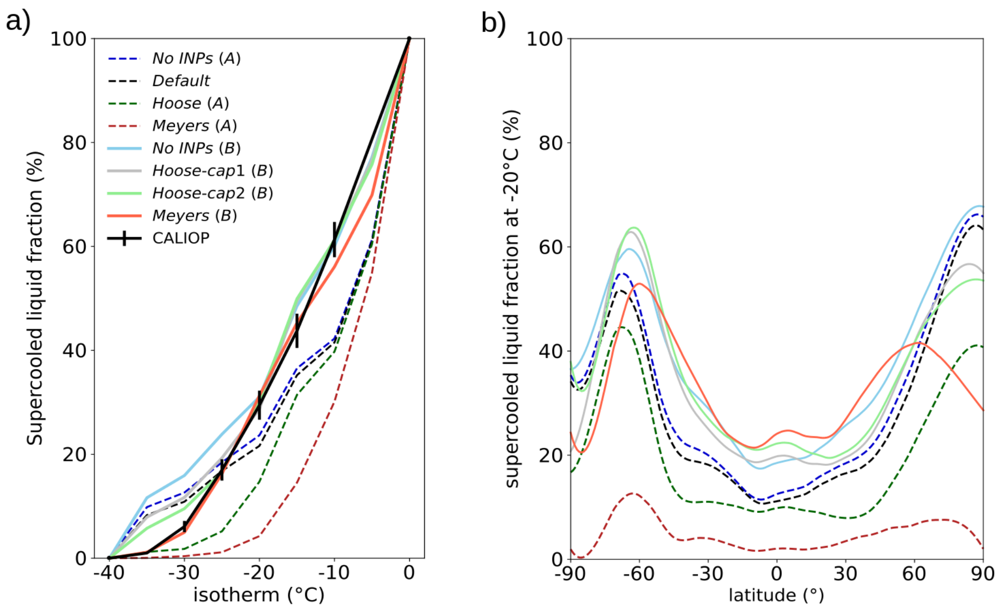

Figure S1 | Supercooled liquid fraction in present-day simulations and CALIOP retrievals, shown by isotherm **(a)** and also by latitude for the -20°C isotherm **(b)**. Only clouds visible to CALIOP are shown, such that clouds under optically thick cloud layers (optical depth τ>3) are ignored. Global mean values are in Table 1. To reduce noise in (a), polynomial smoothing was applied using a window of 20° latitude.

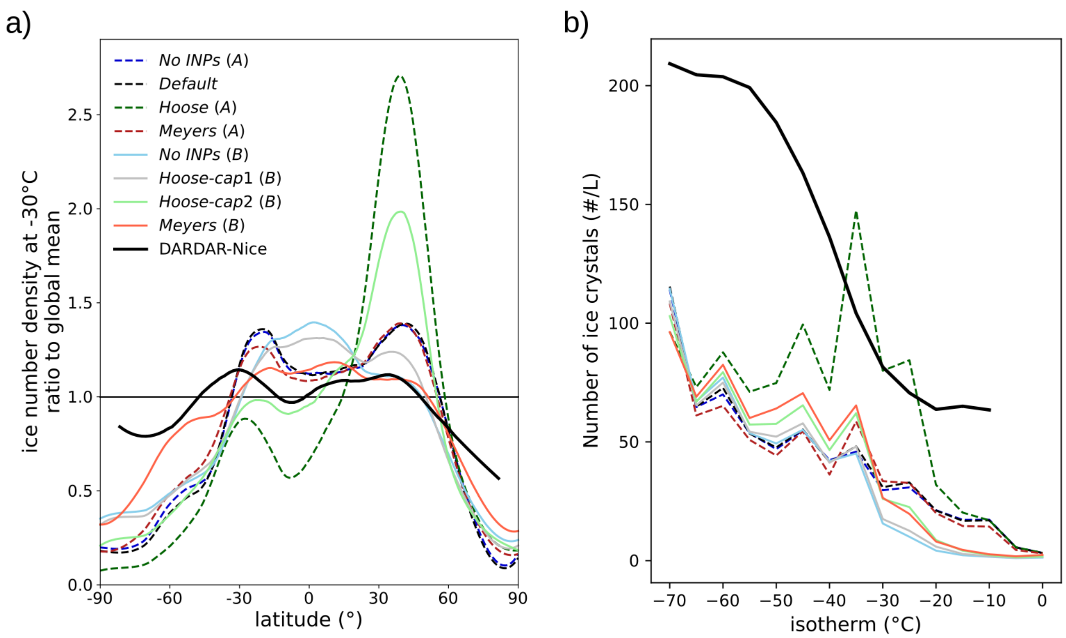

Figure S2 | Ice number density in present-day simulations and DARDAR-Nice satellite retrievals, showing spatial variability of in-cloud ice crystal number near -30°C **(a)** and global mean values across isotherms **(b)**. Data in (a) is normalized by each simulation's global mean value to emphasize differences in spatial structure. This data includes only ice crystals >5μm diameter within ±1°C of each isotherm with gaps of 5°C. As in Fig. S1a, polynomial smoothing was applied to (b) using a window of 20° latitude.

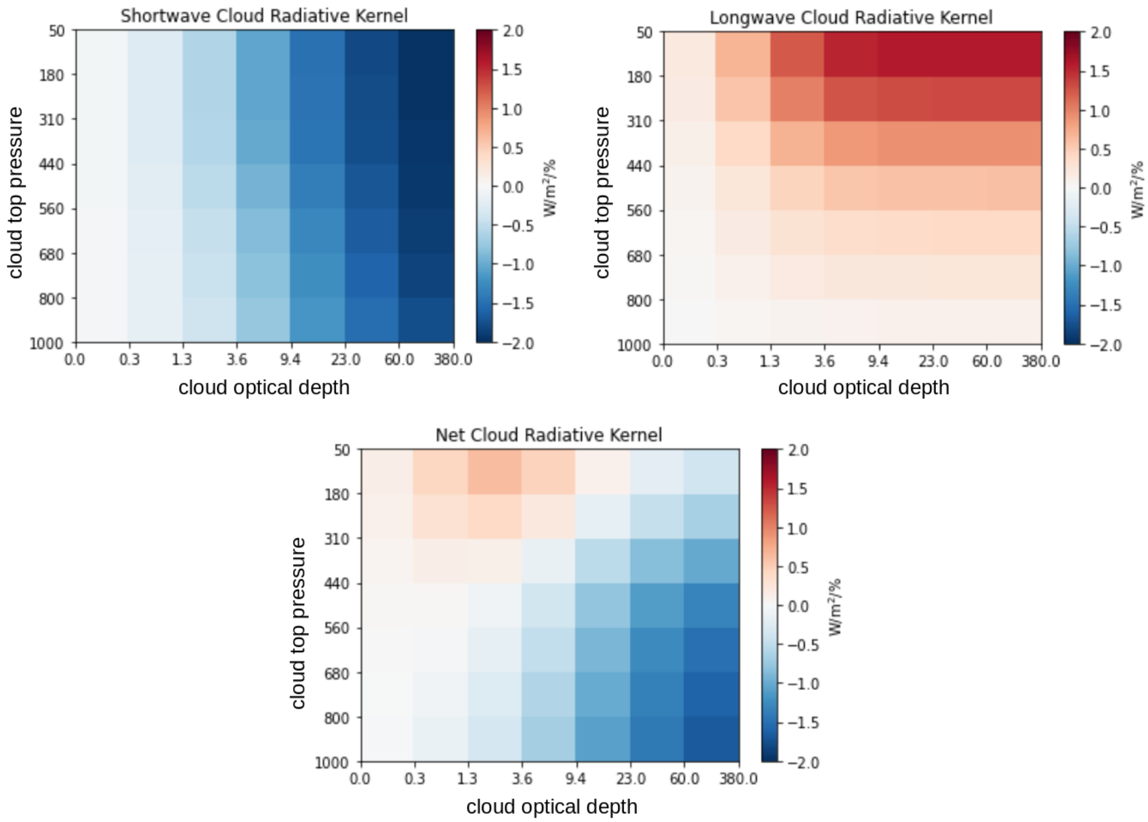

Figure S3 | Radiative kernels used in this study, showing impacts of % changes of each cloud type on shortwave (SW), longwave (LW), and net (SW+LW) radiation. This is as in Fig. 1 of Zelinka et al. (2012), but here SW influence is averaged over surface albedo data from default CESM2.

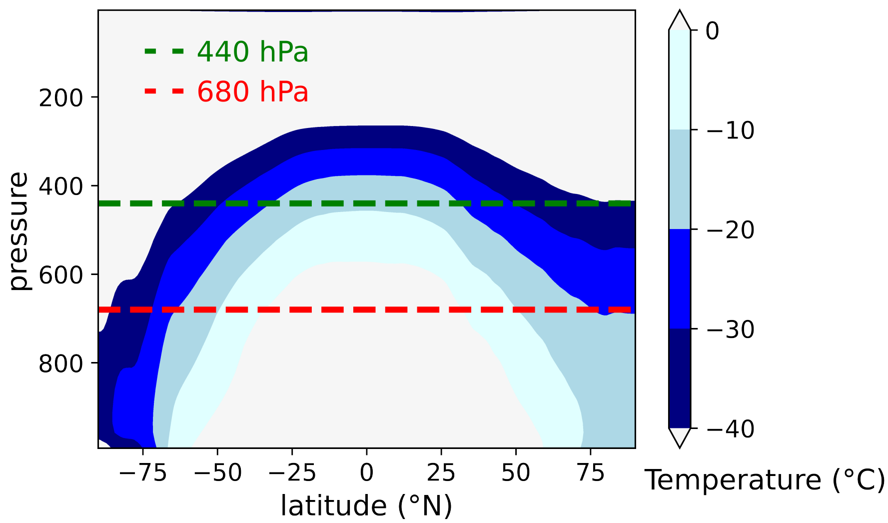

Figure S4 | Locations of isotherms where mixed-phase clouds may exist, showing mean atmospheric temperatures from -40°C to 0°C (blue contours) as a function of latitude and pressure in the *Default* simulation. Isobars at 680 and 440 hPa are shown (red and green dashed lines, respectively) to highlight the boundaries between low, mid-level, and high clouds used in this and other studies.

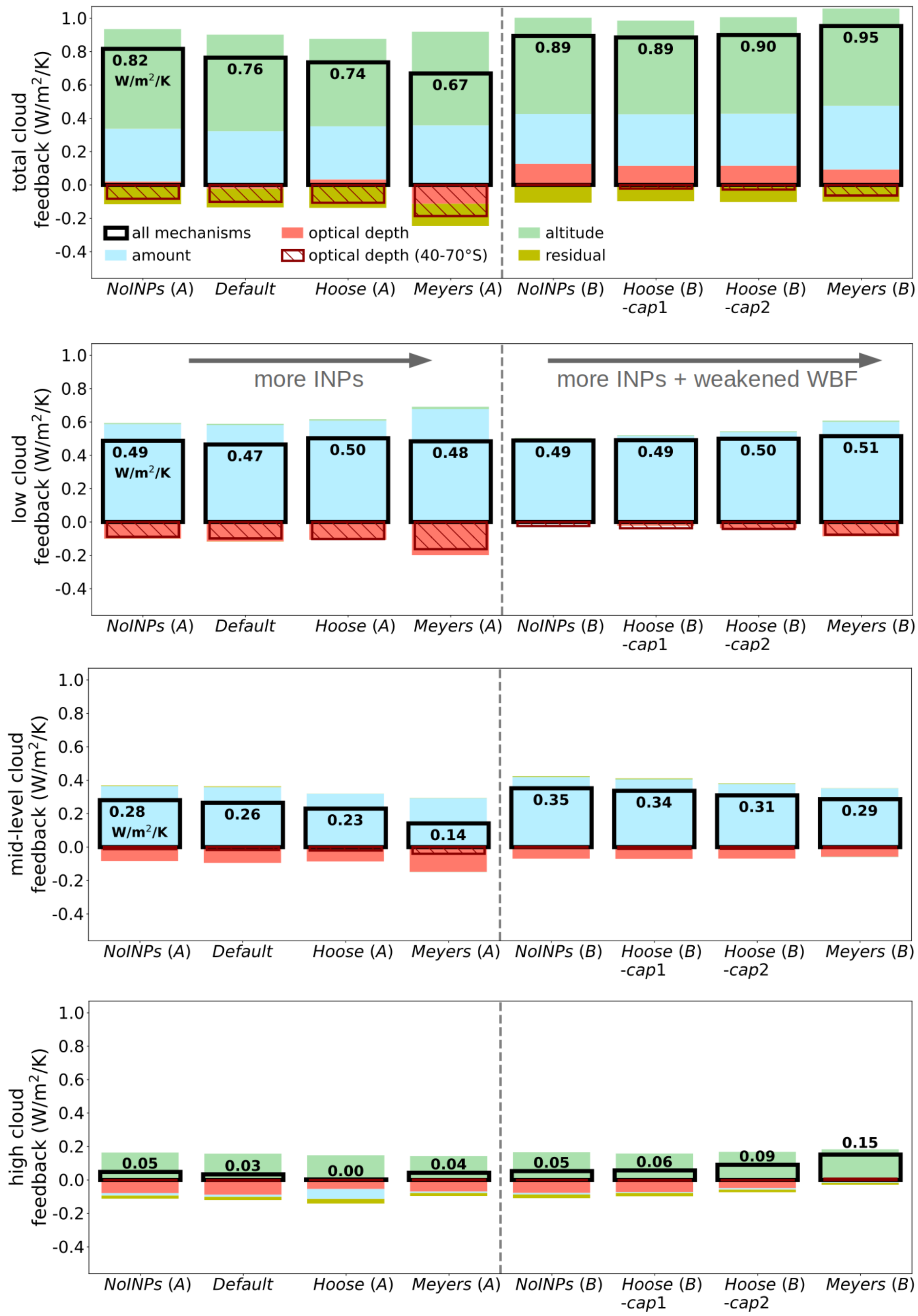

Figure S5 | Cloud feedback strength by mechanism, showing cloud feedbacks in all model experiments, split by individual mechanisms according to the radiative kernel decomposition. All data are global means, aside for the red hatches showing contributions of optical depth feedback over the 40-70°S Southern Ocean region (mean feedback over this region multiplied by its 14.8% share of global surface area). Black hollow bars show sums of all mechanisms, with individual feedback values in W/m^2/K included as black text. For decomposition by both mechanisms and latitude, see Fig. S6.

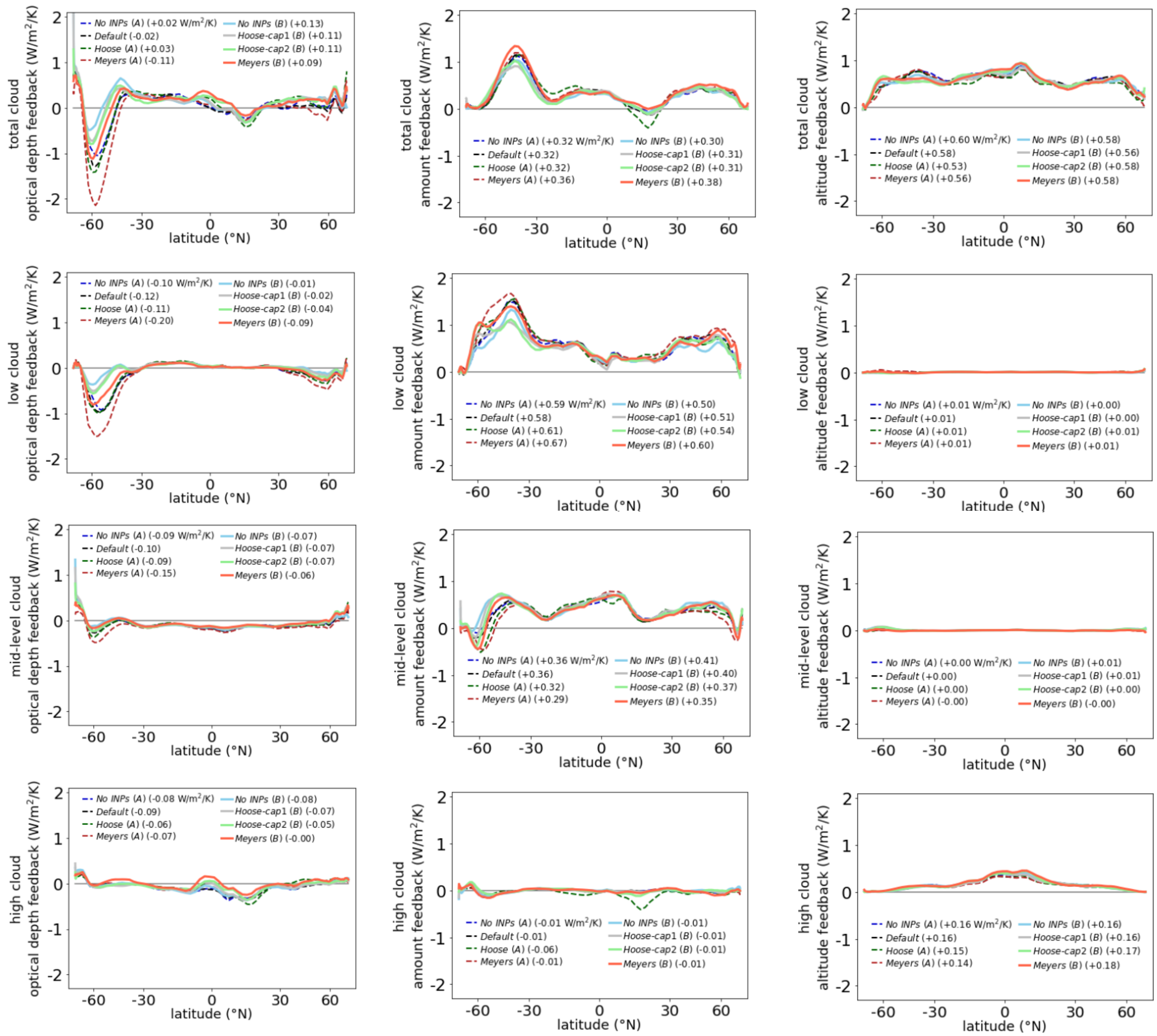

Figure S6 | Cloud feedbacks separated by mechanism in all CESM2 simulations, as in Fig. 2c but further partitioned by mechanism. Global mean feedback values are included in each legend. Note that some mismatch exists between the sum of feedbacks between levels and each unseparated feedback (as is explained in Zelinka et al., 2016), shown here in the top row. Note also that we have not included the residual term that separates the sum of cloud feedback mechanisms from the total cloud feedback.

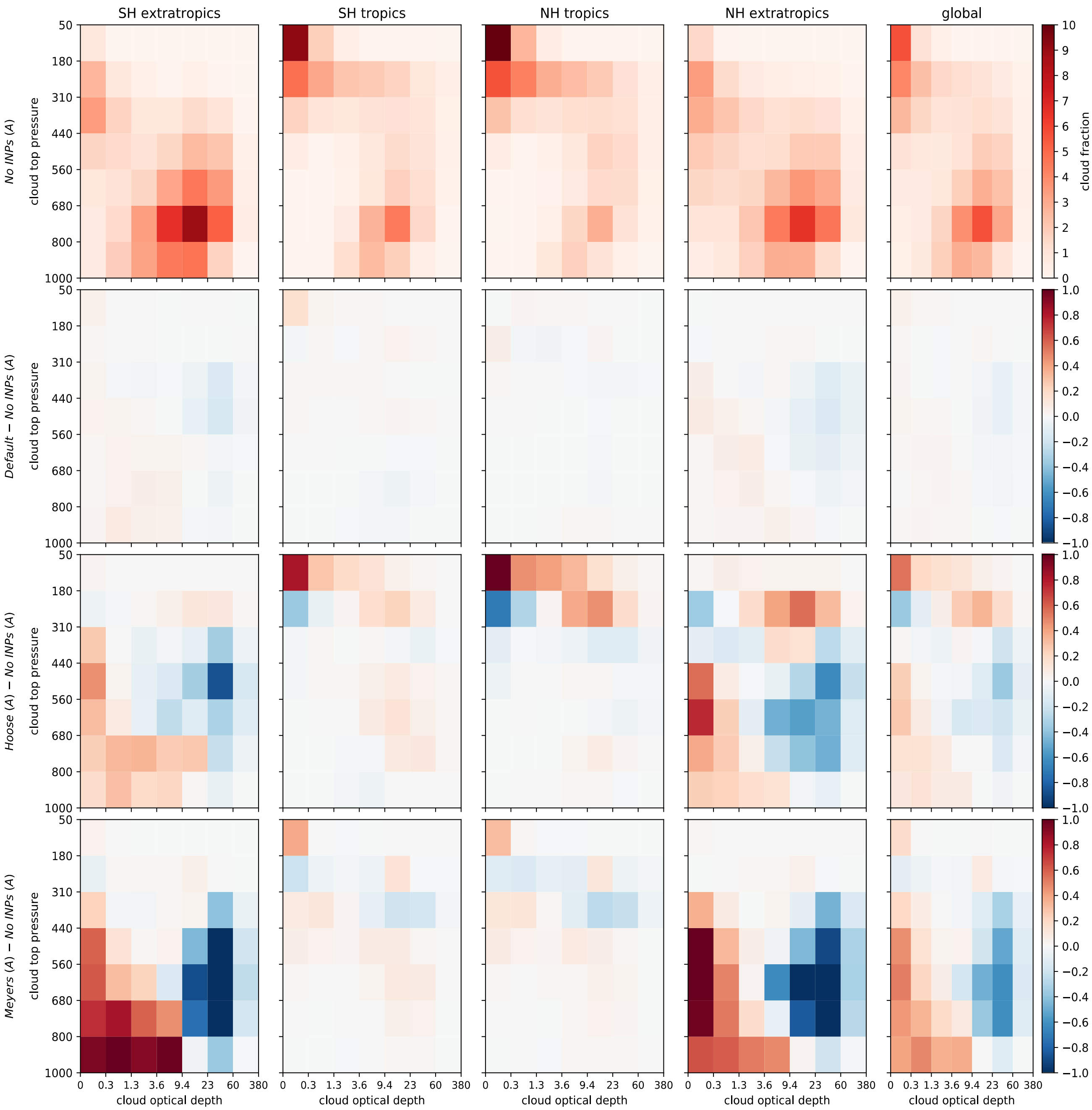

Figure S7 | Cloud types identified in ISCCP simulator cloud histograms, showing changes to prevalence of 49 cloud types. Cloud types are separated by cloud top pressure and optical depth and are shown as standard model output for comparison with ISCCP. Output from *No INPs (A)* is shown directly, while the other unadjusted experiments are shown as difference from these cases for clarity. Note that the difference plots have a color bar ten times stronger than those for *No INPs (A)*. Group B experiments are shown separately in Fig. S8.

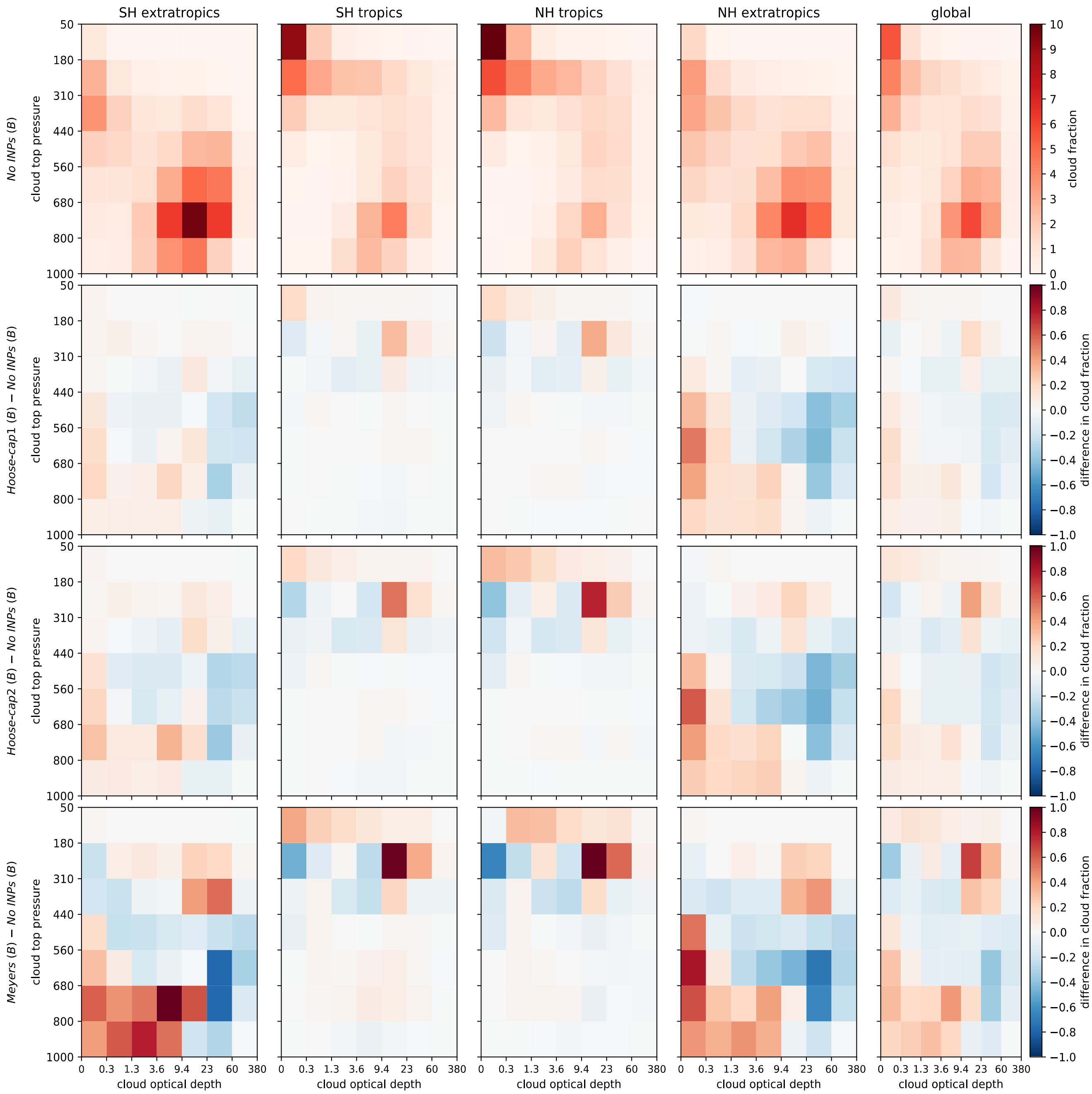

Figure S8 | As in Figure S7 but among the Group B simulations, with experiments here compared to *No INPs (B)*.

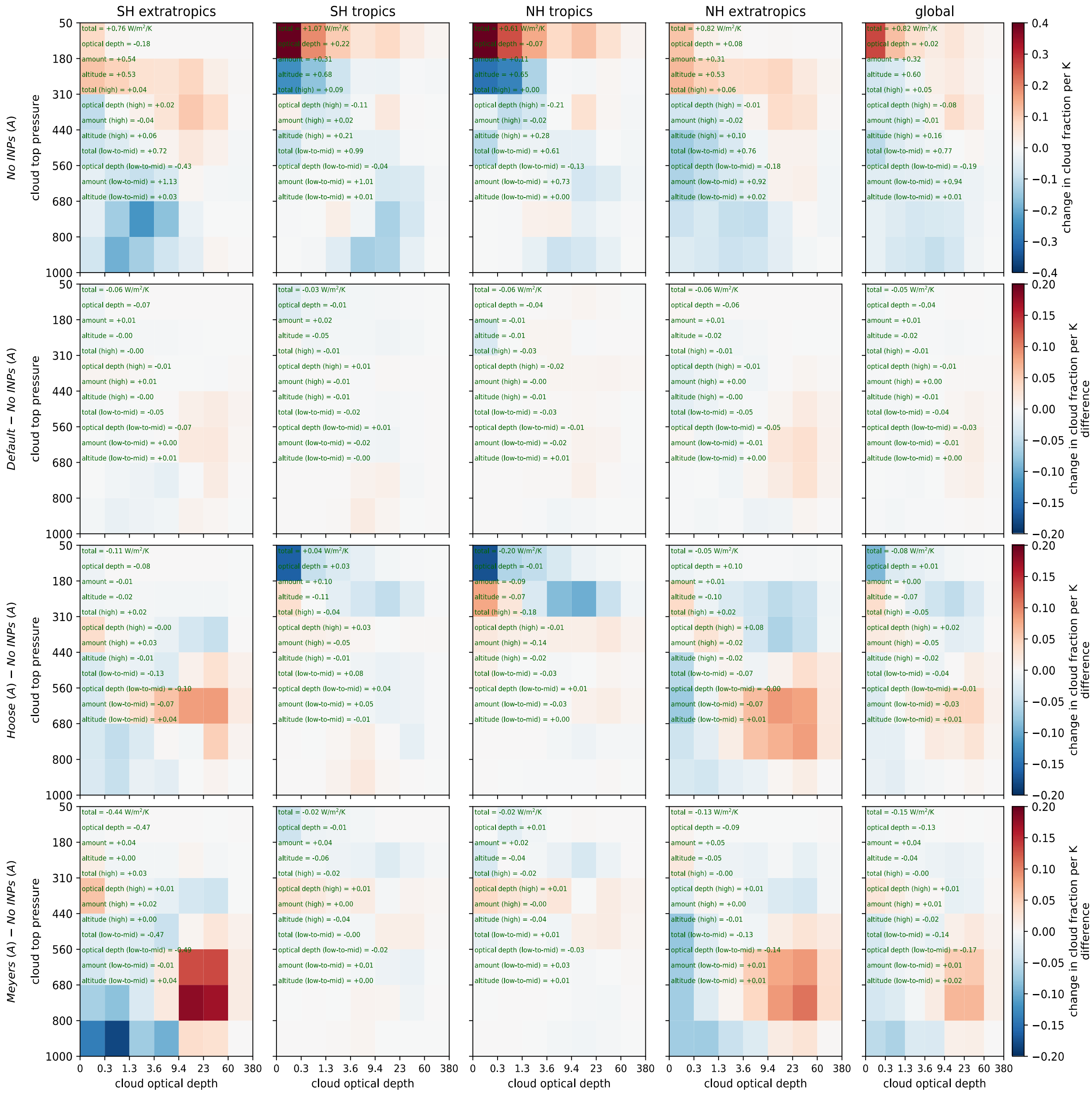

Figure S9 | Cloud changes as warming occurs as identified in ISCCP simulator cloud histograms. Aside for the top row (*No INPs (A, SST+4K) – No INPs (B, present-day)*), the data is shown as four-way differences, i.e. (*experiment*(A, SST+4K) –*experiment(A, present-day)*) – (*No INPs (B, SST+4K) – No INPs (B, present-day)*). Also included on the plots in green text are cloud feedbacks calculated by the kernel method (showing feedback differences compared to *No INPs (A)* below the first row), with low and mid-level clouds grouped together to conserve space. Note that the four-way difference plots have a colorbar twice as strong as the top row. For differences in present-day clouds among the same simulations, see Fig. S7.

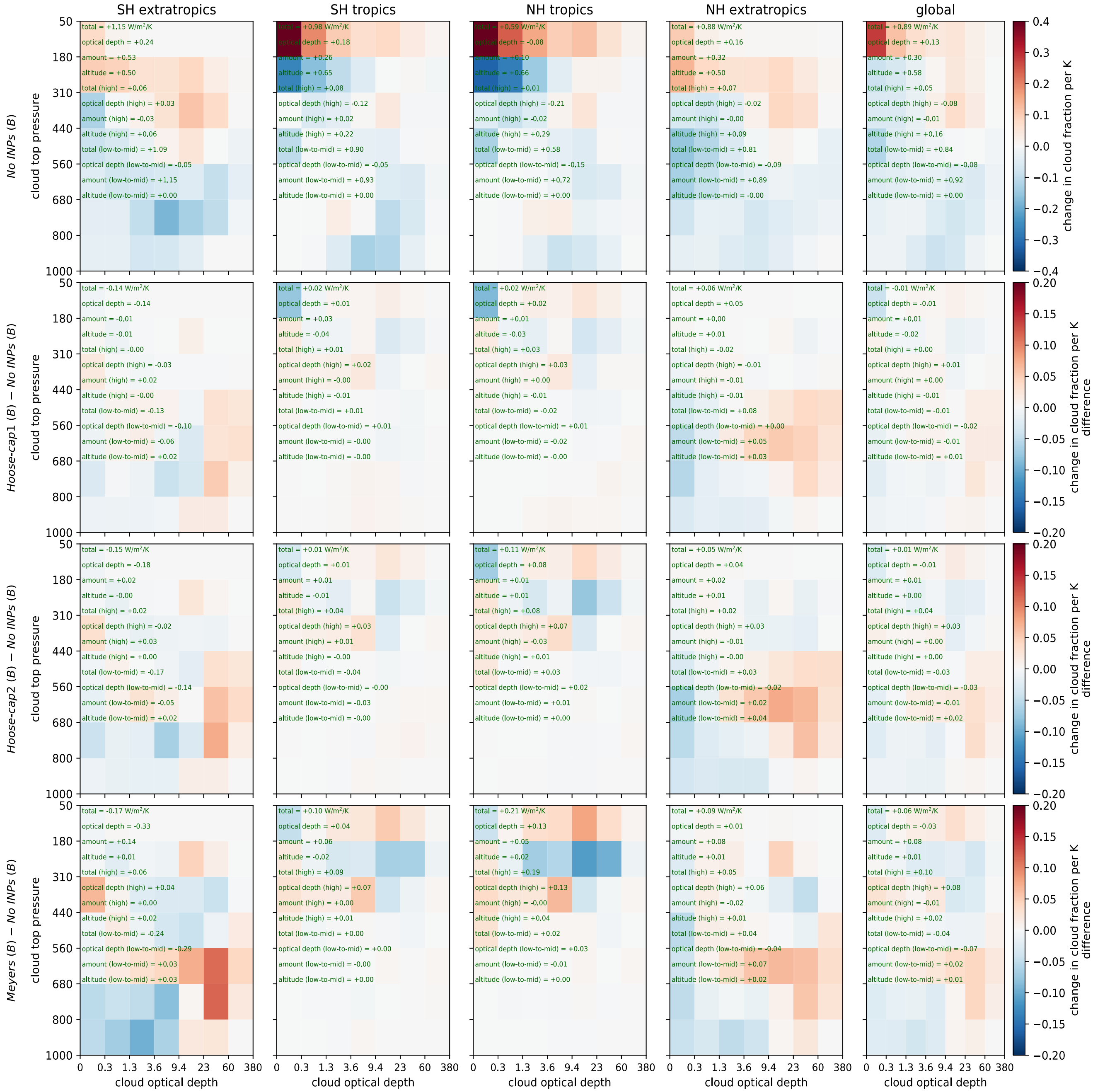

Figure S10 | As in Figure S9 but among the Group B simulations, with experiments being compared to *No INPs (B)*. For differences in present-day clouds among the same experiments, see Fig. S8.

Supplementary References

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