Responses to reviewer comments
The original comments are in *blue italic font*, and our response are in black font.

Reviewer #1:
The authors present a detailed study of cloud property susceptibilities (of liquid water, albedo, and cloud fraction) to droplet numbers in low liquid marine clouds in the NE Atlantic, using new geostationary satellite data. They show diurnal variations in the susceptibilities, with the responses being most negative in the early afternoon – when the widely used Aqua satellite collects data. This could point at a bias in studies quantifying the susceptibilities from this time of day. The study is carefully argued, with the right level of detail, and gives important insights into the responses of clouds and climate to anthropogenic aerosol. But there are a number of major points to be addressed before publication.

We appreciate the positive assessment and the constructive suggestions. Below, we provide a point-by-point response to each comment and details of the modifications made to the text and figures to address these points.

Major comments:
1 Arola et al (2022) showed that even a positive Nd-LWP relationship can give negative correlations when regressing, due to natural variability and retrieval errors. Is this a pertinent potential source of error for your study? They restrict to small retrieval errors (working on MODIS data) and show that the relationships grow weaker. Please test the sensitivity of your results, especially Figure 3 on diurnal variability, to restricting more or less to retrieval uncertainty. Is there a way you can assess the importance of natural variability changing the retrieved slope?

Thank you for your insightful question and for providing the reference paper. As shown in Arola et al. (2022), Feingold et al. (2022), and Zhou and Feingold (2023), small-scale cloud heterogeneity, spatial scale of the cloud organization, or even the N_d retrieval algorithm led to large variabilities in the retrieved LWP and albedo susceptibilities. To account for these factors, aggregation/smoothing of the pixel-level data before the regression is performed (e.g., Feingold et al., 2022; Zhou and Feingold, 2023). In our study, we first smoothed the pixel-level Meteosat retrievals to a $0.25°$ spatial resolution and aggregate all the data over a $10° \times 10°$ domain and 4 years. We have included references to these papers in section 2 to provide a clear explanation of our approach and to emphasize our efforts to mitigate potential biases arising from natural variability. Moreover, before performing the smoothing and aggregation, we removed pixels near cloud edges, which should substantially reduce biases in cloud retrievals.

To assess the sensitivity of our results to Meteosat retrieval uncertainty, given the absence of true values for the retrieved τ and r_e, we manually applied scaling factors of 120% and 80% to the retrieved τ and r_e. As shown in Figures R1 and R2, consistent daytime variation of cloud susceptibility is retained for all four variables. However, the magnitude of change for each variable is sensitive to the retrieved τ and r_e as expected. We have added discussion regarding to the influence of retrieval uncertainty and cloud heterogeneity to the quantified susceptibility in section 2 as follow:

"As found by Arola et al. (2022) and Zhou and Feingold (2023), the retrieved cloud susceptibilities are sensitive to small-scale cloud heterogeneity, the co-variability between cloud properties and N_d, and the spatial scale of cloud organization. To reduce the biases resulting from heterogeneity and co-variability, we first average the 3-km pixel-level cloud retrievals to a regular $0.25° \times 0.25°$ grid for each half-hourly time step. As suggested by Feingold et al (2022), N_d retrieval was performed at pixel-level using Eq. (1), and then averaged to a $0.25°$ resolution." (Lines194-199).

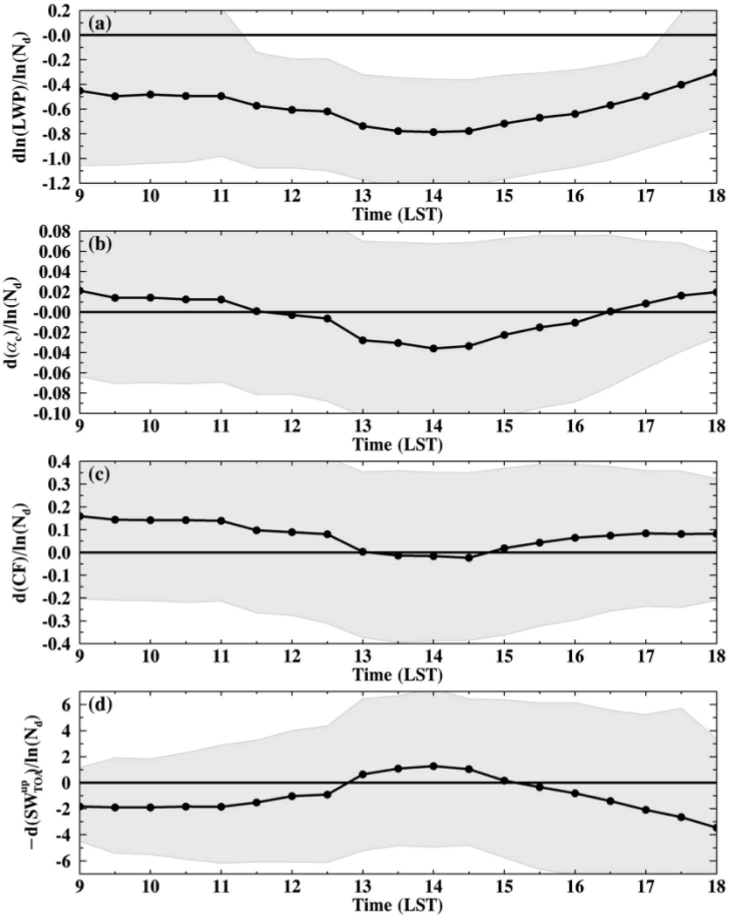

Figure R1. Same as Figure 3, but with retrieved τ and r_e scaled up by 120%.

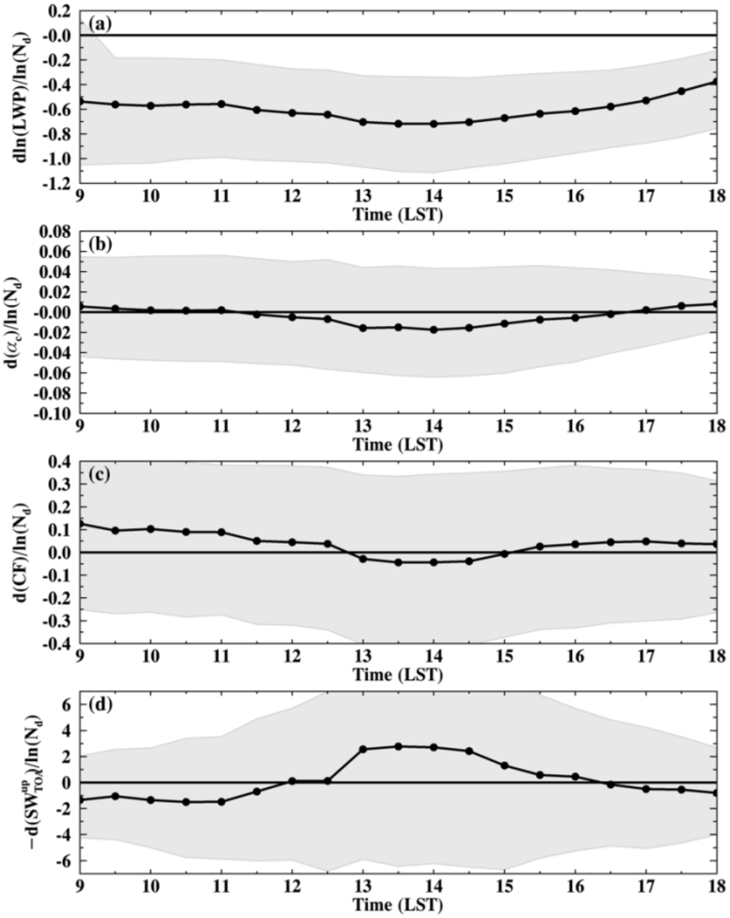

Figure R2. Same as Figure 3, but with retrieved τ and r_e scaled down by 80%.

2. On causality: The retrievals will show causation and covariation: position in the cloud (edge vs. inner part, time variations)? You argue, that we do not expect the meteorological parameters to vary much in the 1deg grid box, but then we expect the same for aerosol concentrations–can we still say that Nd variations are from aerosols alone? This is what is implied in calling the correlations/regression coefficients 'susceptibilities'. To what extent are we able to say that an (e.g. anthropogenic) increase in Nd would lead to a corresponding change in LWP, albedo, CF? Please add a discussion of this.

The scale of heterogeneity and variability in aerosols is typically much smaller than that of meteorological and synoptic conditions. Within a 1° × 1° box, variations in aerosol and N_d can arise from sources like occasional ship emissions, local island emissions, and long-distance transportation. It's acknowledged that, according to the definition of aerosol-cloud interactions in this study, it inevitably comprises the response of cloud properties to N_d perturbation (the targeted signal) and the spatial/temporal covariation between N_d and cloud properties.

To minimize the influence from temporal covariation, we quantify the susceptibility within each time step of satellite observations, similar to Figure 1b verses 1a in Arola et al. (2022). As discussed in the previous comment, to minimize the influence of spatial covariation, we averaged the pixel-level satellite retrievals to a 0.25° spatial resolution and aggregate the data over a $10° \times 10°$ domain for a period of 4 years. However, unlike well-controlled numerical simulations, this observational study alone cannot quantify the contribution from aerosol-cloud interaction versus from covariation or establish causality in the observed relationship. Future studies are discussed in the manuscript to address this challenge.

3. *This becomes particularly important when looking at transitions or 'cloud memory': Does an observed stronger correlation of Nd and LWP in thin clouds which were previously thick mean, that if we now added aerosol, they would dry even more? Or is this more negative correlation because of the processes involved in the thinning of thick clouds? On page 13, regarding your hypothesis 3, you examine the decay of thin clouds, which does not explain the changes. But clouds undergoing a similar 'decay' from thick to thin are shown to have strong susceptibilities. In particular, in line 405, you say that "Similar results are obtained using classification methods based on [different CF thresholds (e.g., from 10% to 30%) or] changes in the mean LWP". To test hypothesis 2 (cloud memory), you use a two-hour LWP change classification. This means that the only difference for testing hypotheses 3&2 is the time scale (30 mins vs. 2h), but it is not immediately obvious why dissipation should not happen more slowly. In short: Could the cloud memory effect (thick-thin) just be dissipation of thick clouds, and the mechanism not cloud memory but covarying effective radii and LWP? To claim 'cloud memory', you need to rule this out.*

Thank you for your insightful question. We have explored different definitions for the dissipation and development of clouds. As shown in Figure S4, when using the change of cloud fraction (CF) as the definition of cloud dissipation and development, non-precipitating thin clouds with increasing or decreasing CF have less negative LWP susceptibilities than clouds with constant CF. Therefore, the decrease of LWP susceptibility from morning to noon is unlikely due to the dissipation and development of thin clouds.

We also tried defining the dissipation and development of clouds using the change of cloud LWP, as shown in Figure R3. Dissipating/developing clouds are defined as clouds with a decrease/increase in LWP greater than 25 gm^{-2} in the past two hours. Results are consistent with different LWP thresholds from 15-45 gm^{-2} (not shown). To compare with the definition of the cloud state transition, a same two-hour time window was applied, which is different from the 30-min time window for Figure S4. As the non-precipitating thick and thin clouds are defined by cloud LWP, results shown in Figure R3 are similar to results shown in Figure 5. Dissipating clouds with decreasing LWP have more negative LWP susceptibility than clouds with constant LWP (Figure R3b). However, differences between dissipating and constant clouds are not statistically significant. This is likely because the decrease of cloud LWP does not distinguish between the transition from non-precipitating thick to thin clouds or the transition from precipitating to non-precipitating thin clouds.

In summary, non-precipitating thin clouds with decreasing LWP retain the "memory" of cloud susceptibility of their previous conditions (Figure 5 and Figure R3). Meanwhile, different transitions in cloud state (e.g., rain → thin vs. thick → thin) show significantly different cloud susceptibilities. Furthermore, the dissipation/development of clouds include change in both cloud LWP and CF, while changes in CF cannot explain the evolution of cloud LWP susceptibility (Figure S4). Therefore, daytime variation of cloud LWP susceptibility is better explained by Hypothesis 1 instead of by Hypothesis 2 in Table 1. We have added a discussion on the different definitions of cloud dissipation and a discussion of Fig. R3 to the manuscript.

"Besides the change in CF, dissipation/development of clouds can be defined by change in LWP. However, as our definition of thin and thick clouds use LWP thresholds, results based on change in LWP are similar to results shown in Fig. 5, but with weaker signal (not shown). This indicates that classification of precipitating verses non-precipitating clouds is necessary in distinguishing cloud responses to N_d perturbations than merely using the LWP threshold." (Lines 501-505).

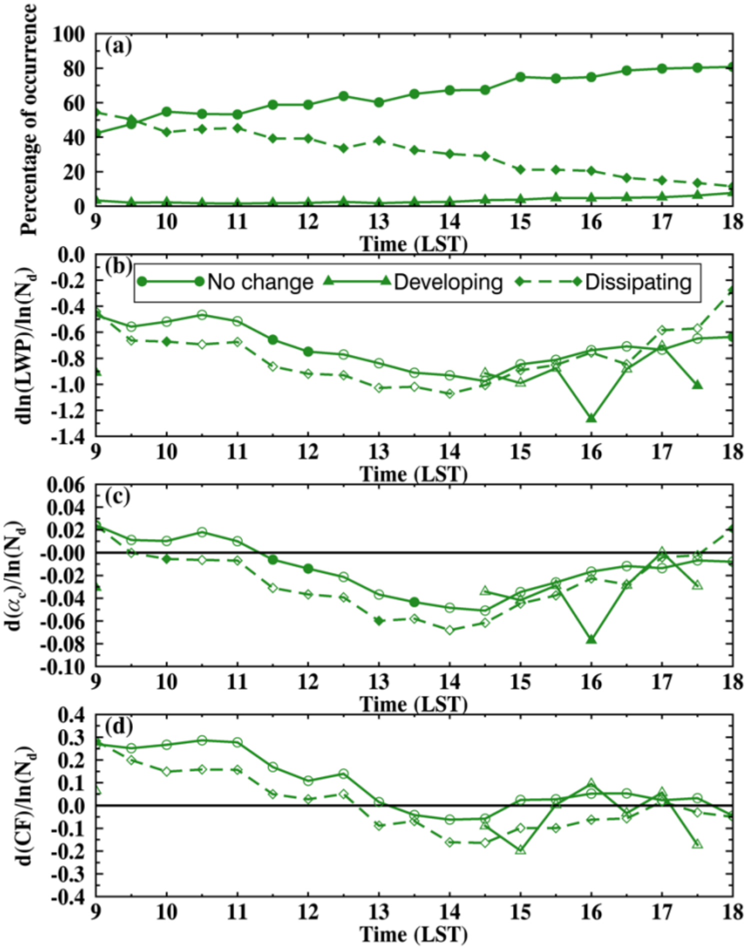

Figure R3. Daytime variation of non-precipitating thin clouds that have small changes in the 1° × 1° mean LWP (No change, solid line with circle symbols), with an increase in LWP (developing, solid line with triangle symbols), and with a decrease in LWP (dissipating, dash line with diamond symbols) within a two-hour window. (a) Percentage of occurrence for the three groups above, (b) cloud LWP susceptibility $(dln(LWP)/dln(N_d))$, (c) cloud albedo susceptibility $(d\alpha_c/dln(N_d))$, and (d) cloud fraction susceptibility $(dCF/dln(N_d))$ for non-precipitating thin clouds. Symbols representing different cloud stages are noted in (b). In (b)-(d), filled markers indicate data points that are significantly different from the other two groups ($p<0.05$). Open markers indicate statistical insignificance.

Specific comments:

-L308-310: "Precipitation acts to stabilize the boundary layer, remove water from cloud top, and reduce the entrainment rate (Sandu et al., 2007, 2008). Precipitation suppression and

entrainment weakening work in concert and result in a net increase in LWP with increasing Nd."
This does not make sense to me. If precipitation tends to reduce the entrainment rate, its suppression strengthens the entrainment rate. So both processes work against each other? Please, could you elaborate on this?

Thank you for pointing out this discrepancy. Because of the influence of precipitation on boundary layer and cloud water, in heavily precipitating clouds, the entrainment rate is smaller compared to that in non-precipitating clouds with similar LWP. With increasing N_d from aerosol perturbations, cloud droplet sizes decrease and suppress precipitation, causing an increase in cloud LWP. Meanwhile, the increased aerosols also enhance cloud top entrainment rate, leading to a decrease in cloud LWP. As the entrainment is weaker in heavily precipitating clouds, the precipitation suppression feedback likely outweighs the entrainment feedback, resulting in a net increase in LWP (e.g., Chen et al., 2014; Toll et al., 2019)). Modifications were made in the tracked changes for clarity: "Therefore, precipitating clouds exhibit smaller entrainment rate than non-precipitating clouds with similar LWP. The increase of LWP from precipitating suppression feedback outweighs the decrease of LWP from entrainment feedback and results in a net increase in LWP (e.g., Chen et al., 2014; Toll et al., 2019)." (Lines: 363-365).

-L334-336: "The opposite signs in LWP and CF susceptibilities for non-precipitating thin clouds cannot be solely explained by the evaporation-entrainment feedback. In the next section, two additional hypotheses regarding the development/dissipation of clouds and the transition of cloud states will be tested."
I agree that the opposite signs in LWP and CF adjustment cannot be explained with evaporation-entrainment. But I do not find an explanation later, after you introduce the hypotheses and conclude susceptibility variations are due to cloud memory. To me, this does not explain the opposing signs. Please include a discussion.

Increases in aerosol decrease cloud drop size and increase cloud drop number concentration near cloud top, which enhance the cloud top radiative cooling rate and the entrainment rate. The enhanced radiative cooling and entrainment induce downdraft and mixing from cloud top (e.g., Xue and Feingold, 2006). These factors contribute to destabilize the boundary layer and facilitate moisture transport from the ocean surface to clouds, thus enhance new cloud formation and extend cloud lifetime (e.g., Christensen et al. 2020). This hypothesis is consistent with and supported by the relative low CF for these clouds (Fig. S2a) and the diurnal variation in LWP susceptibility for non-precipitating thin clouds (Figure 4c). In the morning, the boundary layer is typically shallower and well-mixed with clouds coupled to the surface. Therefore, the CF susceptibility for thin clouds is large positive in the morning, and gradually decreases from morning to noon. However, the near zero CF susceptibility in the afternoon is not supported by this hypothesis. Further analyses and model simulations are needed to better understand the diurnal evolution of aerosols' impact on entrainment rate, boundary layer state, cloud cover and lifetime to explain the observed daytime variation of CF susceptibility for non-precipitating thin clouds. The opposite signs observed in LWP and CF susceptibility suggest that the aerosol indirect effect likely redistributes cloud water horizontally, causing clouds to become thinner and wider. A related discussion has been added to Figure 2 on the daytime mean CF susceptibility and to Figure 5 on the daytime variation of CF susceptibility as follow.

"A possible explanation for the increased CF is the enhanced cloud top radiative cooling rate help to mix the boundary layer facilitate moisture transport from the ocean surface to cloud, and therefore favor new cloud formation and extend cloud lifetime (e.g., Christensen et al. 2020). This hypothesis is consistent with and supported by the relative low CF for these clouds (Fig. S2a) and the diurnal variation in LWP susceptibility for non-precipitating thin clouds, which will be discussed in the next section. The opposite signs of LWP and CF susceptibilities indicate that the AIE might redistribute cloud water horizontally and make the thin clouds thinner and wider." (Lines 405-410).

"As the CF susceptibility for thin clouds transitioned from precipitating clouds and thick clouds greatly decrease from morning to noon, the CF susceptibility for thin clouds decrease from large positive to near zero from morning to noon (Fig. 4c). Another possible explanation on the evolution of CF susceptibility is the influence of aerosols on boundary layer mixing and the evolution of boundary layer from morning to noon. The enhanced entrainment rate and radiative cooling rate from N_d perturbations help to destabilize the boundary layer and transport moisture from the ocean surface to clouds, which facilitate new cloud formation. As the boundary layer is typically well mixed in the morning with clouds coupled to the surface, this impact is strongest in the morning and gradually decrease from morning to noon. In the afternoon, on the other hand, thin clouds transition from all three states have near-zero CF responses to N_d perturbations, which cannot be explained by the hypothesis above. Further analyses and model simulations are needed to better understand the diurnal evolution of aerosols' impact on entrainment rate, boundary layer state, cloud cover and lifetime to explain the observed daytime variation of CF susceptibility for non-precipitating thin clouds." (Lines 549-560)

-L368: "The diurnal variation of cloud susceptibility is statistically significant at a 95% confidence level based on a student's t-test." Please elaborate: What are the variables/means that are compared here, with what variability?

We compared all four cloud susceptibilities at different times of the day. The results indicate that cloud susceptibilities in the morning and evening exhibit a statistically significant difference compared to those at noon, established at a 95% confidence level. We also did a trend analysis of the daytime mean value of the cloud susceptibility, which is statistically significant. The high variability in cloud susceptibility includes both spatial and temporal variability, which highlights the complex interplay between synoptic conditions that varies diurnally and cloud states in the ENA region.

-Figure 2: What is the variability (e.g. the standard error of the mean) in the averaged 1x1deg susceptibilities for each LWP-Nd bin? Can you state which average susceptibilities are significant? Why are some squares missing in some panels but not in others?

The variability of the cloud susceptibilities for each N_d and LWP bin is shown in Figure R4. The difference in cloud susceptibility for different cloud states are statistically significant for all four variables. The blank bins in Figure 2 are bins with sample number smaller than 100. Discussion on the variability of the 1° cloud susceptibilities have been added to section 3.2 in the manuscript.

"The variability of LWP susceptibility in different LWP-N_d bins vary between 0.4 to 1.2 (not shown), while the LWP susceptibilities for precipitating clouds are statistically significant than other two cloud states at a 95% confidence level." (Lines 354-355)

"The variability in the overall 1° α_c and CF susceptibilities range between 0.05-0.15 and 0.3-0.6, respectively (not shown). The α_c and CF susceptibilities for precipitating clouds are statistically significant than other two cloud states at a 95% confidence level." (Lines 375-376)

"Non -precipitating thick clouds exhibit the greatest variabilities in their LWP and α_c susceptibilities in the three cloud states. The 1° LWP and α_c susceptibilities vary between 1.0-2.4 and 0.2-0.25, respectively (not shown). Due to the enhanced entrainment and evaporation, the mean CF mostly decreases with increasing N_d, with mean CF susceptibilities ranging from -0.1 to $+0.04\ ln(N_d)^{-1}$ (Fig. 2c). Variability in CF susceptibilities for non-precipitating thick clouds is larger than that for precipitating clouds and smaller than the non-precipitating thin clouds (not shown)." (Lines 389-393)

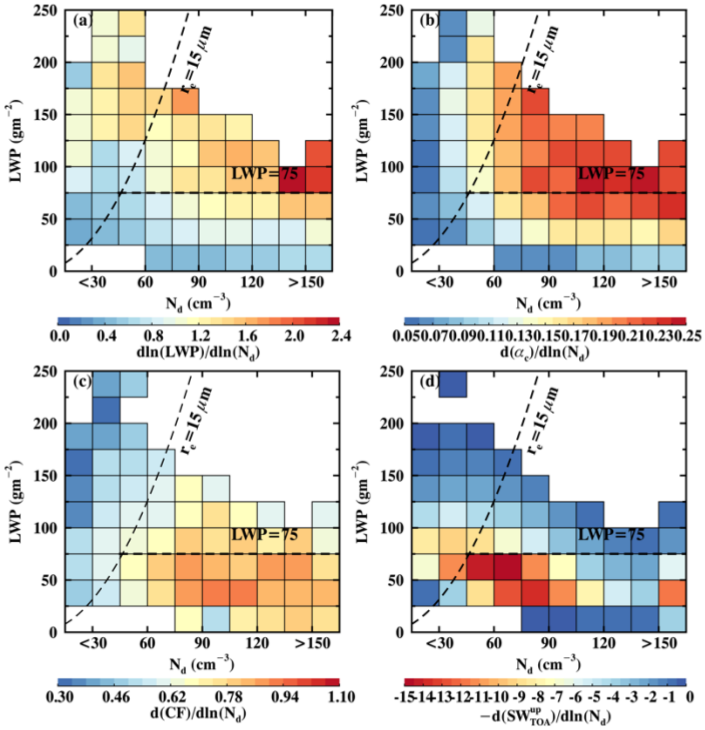

Figure R4. Difference between the upper 75th percentile and the lower 25th percentile of cloud susceptibilities for different N_d and LWP bins during the daytime. (a) cloud LWP susceptibility ($dln(LWP)/dln(N_d)$), (b) cloud albedo susceptibility ($d\alpha_c/dln(N_d)$), (c) cloud fraction susceptibility ($dCF/dln(N_d)$), (d) cloud shortwave susceptibility ($-dSW_{TOA}^{up}/dln(N_d)$) weighted by the frequency of occurrence of samples of each bin.

- L616: "The less stable condition over the studied region leads to a deeper boundary layer, deeper clouds, and a stronger entrainment rate at the cloud top, all of which may cause a more negative LWP susceptibility"

To me, this raises the question how much can we extrapolate to all year liquid marine clouds from July data. If the susceptibility is a function of boundary layer depth (which it should be), then it will change with changing SSTs over the seasonal cycle. Please discuss.

The LWP and albedo susceptibilities should be less negative and more positive, respectively, in other seasons compared to summer in the ENA region. The boundary layer is shallower and less stable in winter, spring, autumn than in summer, and there are more cumuli, convective and heavily precipitating clouds and less stratiform clouds in the boundary layer. All of these factors lead to larger LWP and albedo susceptibilities. The annual mean values should be close to Zhang and Feingold (2023) study. The following discussion has been added to the paper: "This is due to different seasons and study regions between our and their studies. The summer boundary layer in the ENA region is deeper and less stable with higher cloud tops (e.g., Klein and Hartmann, 1993; Ding et al., 2021; King et al., 2013) compared to the NE Pacific in Zhang et al. (2022) and the NE Atlantic region in Zhang and Feingold (2023)." (Lines 750-753)

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