Response to Reviewer #2

We appreciate your time and effort in thoroughly reviewing our manuscript. We are truly grateful for your constructive comments and insightful suggestions, which encourage and help us to improve the manuscript. We have revised the manuscript carefully based on your comments. In the response below, your comments are provided in black text and our responses are provided in blue text.

Response:

SUMMARY

The authors present a comparative study of aircraft in situ aerosol and cloud microphysical measurements of liquid phase boundary layer (BL) clouds from two different regions: over the Eastern North Atlantic (ENA) region near the Azores and the Southern Ocean (SO) in an area spanning south of Tasmania. The ENA measurements are from the summer and winter seasons, while the SO measurements are from summertime only. The overall conclusions are that:

1. Clouds across different regions and seasons have differing collections of microphysical properties
2. These differing microphysical regimes exhibit different susceptibilities to aerosol perturbations
3. Drizzle has a big impact on the BL CCN budget
4. Turbulence plays a leading role in enhanced precipitation seen in the ENA winter regime

The primary data analyzed in the study are aerosol and cloud microphysical properties averaged over all full cloud soundings of warm BL clouds during each campaign. These properties include number concentration (total, modal, fully size-resolved, etc.), measures of drop size distribution (DSD) width, effective radius/mean diameter, liquid water content, sedimentation rate, etc.

Many (if not most) of these observations have been analyzed in other recent studies, and indeed many of the conclusions reached by the authors are "consistent with" (or similar language) the results of these other papers. The frequency with which conclusions are followed by such qualifiers gives the impression that there is not much new added by the manuscript. It would be more effective to give a condensed overview of in situ work on the ACE-ENA and SOCRATES campaigns in which you lay out what has already been done. Then in the results section, devote a subsection (or a couple paragraphs, whatever) to explain how your work fits with what's already been published. This would be easier to digest than the piecemeal and repetitive referencing in the manuscript’s current state. I do see novelty in the specific focus on interactions among aerosol, clouds and precipitation (abbreviated ACI or ACPI, depending on the context), which to my knowledge have not been addressed in the literature for either field campaign (ACE-ENA and SOCRATES) analyzed.

I have serious concerns about the lack of justification for combining analysis of the field campaigns used here as well as the scientific reasoning leading to the point that turbulence- enhanced collision-coalescence explains more intense drizzle during one campaign (ACE-ENA winter) versus the other two. In fact, I think the differences in precipitation attributed to turbulence can be explained much more simply without appealing to turbulence-microphysics interactions *at all* (see 3rd major comment below). While there is clearly a body of legitimate analysis presented in the study, I recommend the manuscript either undergo VERY major revisions or that it be rejected so the authors have sufficient time to rewrite the manuscript before resubmitting it.

Thanks for the thorough assessment of our manuscript, we have tried our best to carefully consider and address your comments and suggestions.

We have reconstructed our discussion to review the previous works on ACE-ENA and SOCRATES in the introduction, and our results in comparison with the previous study in the summary and conclusion section. Please refer to the response to general comment 1 and the revised manuscript.

GENERAL COMMENTS

1. **The unifying thread tying together measurements from the two regions is not clear to me, and I ask that the authors further emphasize/clarify the scientific motivation for combining the campaigns**. This would give a stronger basis for communicating the significance of your results. As it stands, there are no previously unexplored commonalities across the 3 campaigns. Yes, cloud effective radius increases with height for all the campaigns; yes, drizzle mean diameter increases from cloud top to base – these results (among others) are expected, and simply demonstrate that atmospheric physics as we know it isn’t completely “broken.” But beyond that, what purpose does this comparison serve? The aerosol and meteorology driving the clouds in each regime are quite different, so it’s somewhat of a trivial conclusion that the microphysical properties differ as well.

The argument that "SOCRATES and ACE-ENA both took place in the midlatitudes, so they're directly comparable" is insufficient. The SO region sampled by SOCRATES is more consistently impacted by midlatitude cyclone systems than ENA during summer, which is more often dominated by the nearby Azores high (i.e., ENA is borderline subtropical during summer). In addition, I do not buy that these aircraft campaigns can be taken as "representative" samples of their respective latitude bands/ocean basins; Mechem et al. (2018) show significant interannual variability in the synoptic conditions experienced at the ENA site, and the summer ACE-ENA IOP was characterized by anomalously low BL heights *and* substantial BL decoupling.

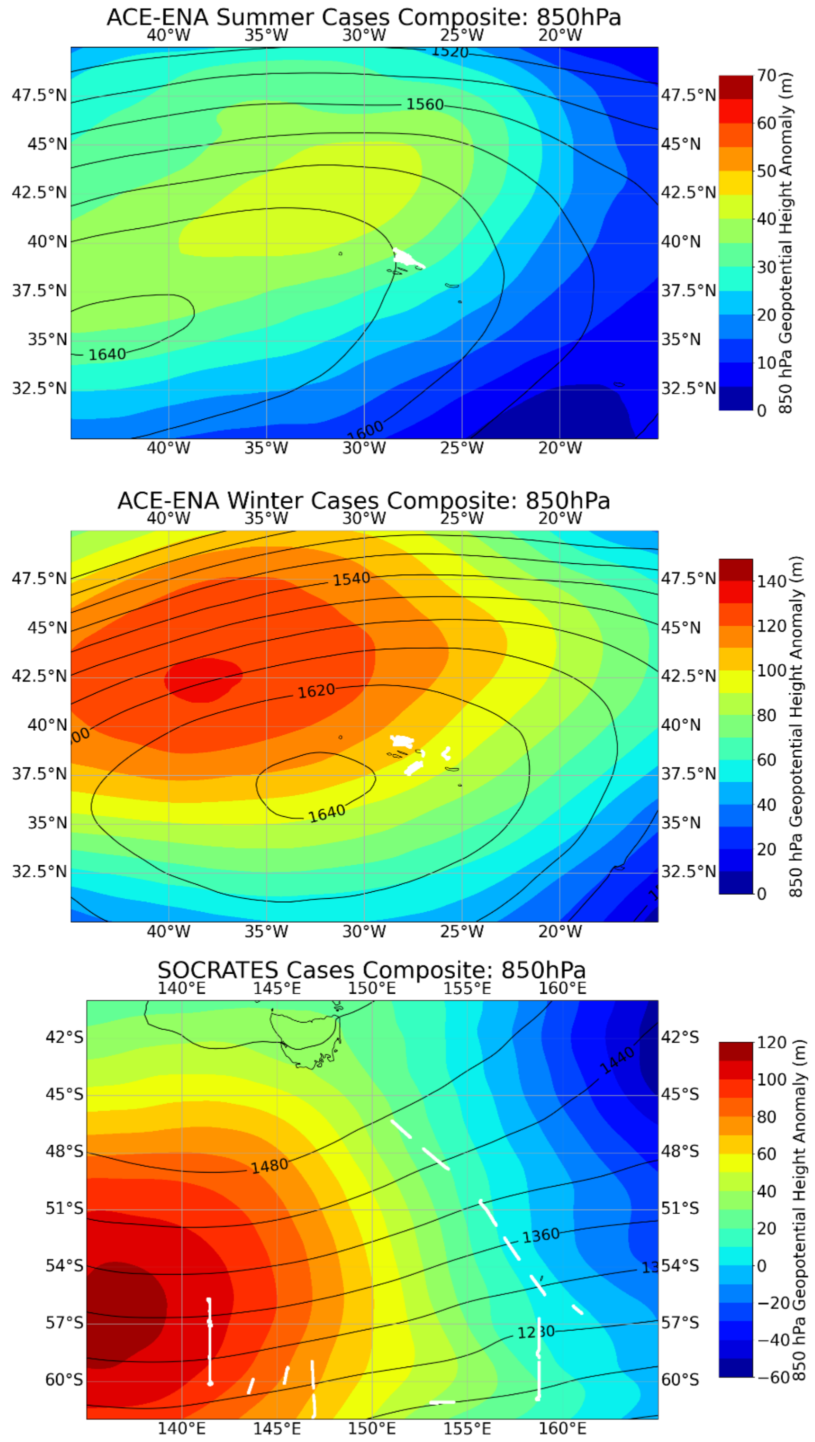
To demonstrate the feasibility and justification of combining these two campaigns, we calculated the composite of the selected case periods during the ACE-ENA and SOCRATES using the hourly ERA5 reanalysis, as shown in Figure S1 below. The composite 850 hPa geopotential height is denoted by black contours, and the height anomaly from the 20-year climatology (2000-2020) is portrayed by the shaded area.

The discussion in the introduction has been modified to better motivate our study, as follows:

'The Eastern North Atlantic (ENA) stands as a desirable region for exploring MBL clouds in the mid-latitude, with Graciosa Island in the Azores (39.09°N, 28.03°W) representing a focal point for such studies. Located between the mid-latitude and subtropical climate zones, Graciosa is subject to the meteorological influence of both the Icelandic Low and the Azores High, and the influence of aerosols ranging from pristine marine air masses to those heavily influenced by continental emissions from North America and Northern Europe (Logan et al., 2014; Wood et al., 2015; Wang et al., 2020). Addressing the need for sustained research into the MBL clouds, the recent Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) aircraft campaign (J. Wang et al., 2022) were conducted in the summer (June and July) 2017 (ACEENA Sum) and winter (January and February) 2018 (ACEENA Win). During these two intensive operation periods (IOPs) of ACE-ENA, the research aircraft accrued abundant in-situ measurements of aerosols, clouds, and drizzle properties, providing invaluable resources for studying the ACI and ACPI processes. During the summer, the Azores is located at the eastern part of the high-pressure system, while during the winter, the center of the Azores high shifts to the eastern Atlantic and is primarily located directly over the Azores (Mechem et al., 2018; J. Wang et al., 2022). Furthermore, both summer and winter IOPs of ACE-ENA are featured with anomalous stronger high-pressure systems, compared to the 20-year climatology as shown in Figure S1. This meteorological pattern is favorable to the prevailing and persistent stratocumulus clouds observed during the ACE-ENA, especially for the winter IOP, where the enhanced large-scale subsidence would lead to a deeper stratocumulus-topped MBL (Rémillard and Tselioudis, 2015; Jensen et al., 2021). The ACE-ENA summer IOP is characterized by anomalously low MBL heights and substantial MBL decoupling (Miller et al., 2021; J. Wang et al., 2022), while the winter IOP is featured with prevalent precipitation-generated cold pools, where evaporative cooling alters the thermodynamical structure of the MBL, sustains and enhances turbulence mixing, hence contributes to dynamical perturbations that can influence the behavior of the MBL (Terai and Wood, 2013; Zuidema et al., 2017; Jenson et al., 2021; J. Wang et al., 2022). Over the recent years, many observational studies, based on the ACE-ENA data, have focused on the seasonal contrasts of the aerosol distributions and sources (Y. Wang et al., 2021b; Zawadowicz et al., 2021), the cloud and drizzle microphysics vertical distributions (Wu et al., 2020a; Zheng et al., 2022b), as well as the impacts of MBL conditions on the cloud structure and morphology (Jensen et al., 2021). However, they seldom analyze the comprehensive interactions between aerosol, clouds and precipitation.

Over the Southern Ocean (SO), the Southern Ocean Clouds Radiation Aerosol Transport Experimental Study (SOCRATES) field campaign (McFarquhar et al., 2021) was conducted during the austral summer (January and February 2018), which marks another valuable piece of the MBL cloud research. The SO, being one of the cloudiest regions globally, is predominantly influenced by naturally produced aerosols originating from oceanic sources due to its remoteness, where the anthropogenic and biomass burning aerosols exert minimal influence over the region (McCoy et al., 2021; Sanchez et al., 2021; Twohy et al., 2021; Zhang et al., 2023). The aerosol budget in this region is primarily shaped by biological aerosols, which nucleate from the oxidation products of dimethyl sulfide (DMS) emissions, as well as by sea spray aerosols. Hence, the SO provides an unparalleled natural laboratory for discerning the influence of these natural aerosol emissions on the MBL clouds under a pre-industrial natural environment. The summertime SO region, particularly near the SOCRATES focus area, is characterized by more frequently closed-cell mesoscale cellular convection structures (Danker et al., 2022; Lang et al., 2022). Furthermore, the MBL clouds over the SO predominantly consist of supercooled liquid water droplets, which coexist with mixed- and ice-phase processes (Y. Wang et al., 2021a; Xi et al., 2022), while the precipitation phases are examined to be primarily dominated by liquid hydrometeors (Tansey et al., 2022; Kang et al., 2024). The in-situ measurements collected from SOCRATES have cultivated numerous studies on aerosols, clouds, and precipitation over the SO using both in-situ measurements and model simulations (McCoy et al., 2020; Altas et al., 2021; D'Alessandro et al., 2021), and provides an opportunity to study the liquid cloud processes under a colder nature. As shown in Figure S1c, compositely speaking, the SOCRATES cloud cases used in this study are located ahead of the anomaly-stronger thermal ridge and behind the thermal trough, providing a set up favorable to the closed cellular MBL cloud structures (McCoy et al., 2017; Lang et al., 2022). While the region of selected SOCRATES cloud cases crosses a larger latitudinal zone and is under more consistent influence of mid-latitude cyclone systems than over the ACE-ENA region, the cloud sampling periods used in this study majority reside in the closed-cell MBL stratocumulus decks.

The cloud cases selected from the ACE-ENA and SOCRATES share similar cloud morphology (stratocumulus) while experiencing different aerosol sources and meteorological conditions. Using a synergistic approach to compare data from these different field campaigns can provide valuable insights to the community regarding the functioning physical processes of the interactions between aerosols, clouds, and precipitation under the influence of different MBL dynamic and thermodynamic conditions. This study targets the similarities and differences in the MBL aerosol, cloud, and drizzle properties, their distribution and evolution, and more appealingly, the ACIs and ACPIs between the two campaigns.’



ACE-ENA Summer Cases Composite: 850hPa

ACE-ENA Winter Cases Composite: 850hPa

SOCRATES Cases Composite: 850hPa

Figure S1. The selected-cases 850 hPa geopotential height composite (black contours), and the height anomaly from (shaded area) the 20-year climatology (2000-2020), for a) ACE-ENA summer, b) ACE-ENA winter, and c) SOCRATES. White dots denote the aircraft flight tracks for the selected cases.

2. The authors go to some length to justify their assertion that turbulence-enhanced collision-coalescence is the reason for stronger precipitation during winter at ENA, but the evidence given does not prove their hypothesis. **The discussion of TKE is illustrative but quantitatively insufficient.** For one, very few details are given on how the velocity perturbations are calculated; for example, what is the integral length scale obtained with a 10 s moving window (i.e., are you capturing the inertial subrange)? Is it the same for ACE-ENA and SOCRATES? (no) Is any window function applied or is this a simple "boxcar" moving average? Is 1 Hz data used or did you analyze high-rate data? Is "high-rate" the same for ACE-ENA and SOCRATES? (no) In addition, TKE is not the relevant quantity for evaluating turbulent enhancement of collisional growth; rather, it is the TKE dissipation rate ϵ that is used in parameterizations (e.g., Grabowski and Wang 2013 as referenced in the manuscript). For another, $\epsilon \sim O(10^{-4} m^2/s^2)$ in stratiform BL clouds while in shallow cumulus it is about an order of magnitude higher. Based on the sampling goals of both ACE-ENA and SOCRATES, mostly stratiform clouds were sampled, suggesting generally low turbulence intensities. A past modeling study on the feasibility of turbulence to overcome the "warm rain bottleneck" and accelerate drizzle formation via collision- coalescence enhancement in subtropical marine stratocumulus (Sc) showed a minor impact (Witte et al. 2019, doi: doi: 10.1175/MWR-D-18-0242.1) – why is a different answer expected in the same cloud dynamical regime? Finally, cold pools are a dynamical forcing mechanism more prevalent during ACE-ENA winter than either of the other 2 campaigns; this is not mentioned at all.

   For you to continue pushing the line of reasoning that turbulence directly *causes* stronger precipitation, at very minimum you need to demonstrate that ϵ is substantially stronger for the ACE-ENA winter campaign than both typical marine Sc *and* SOCRATES/ACE-ENA summer (i.e., much greater than the 30-40% difference in mean cloud-top TKE shown). If you are unable to demonstrate this, I cannot support the heavy reliance on turbulence-enhanced collision-coalescence peppered throughout the manuscript. If you move forward with quantifying, please explicitly detail your approach in the methodology section – I recommend Siebert et al. (2010, 10.1175/2009JAS3200.1) or Waclawczyk et al. (2017, doi: 10.5194/amt-10-4573-2017) as starting points for developing your own analysis.

Thanks for the great suggestions. After carefully considering and examining this aspect, we found that the 1Hz used in this study is not sufficient to capture the small-scale turbulent structures. Thus, we have added the method description, and the following discussion of limitations:

‘The turbulent perturbations of vertical ($\overline{w'^2}$) and horizontal ($\overline{u'^2}$ and $\overline{v'^2}$) components are calculated as the simple moving variance in a 10s window centered at the measurement time, without window weighting function, using 1Hz data for all three IOPs. The *w* data is confined to an absolute aircraft roll angle of less than 5° (Cooper et al., 2016). Given the average aircraft ground speed of ~140 m/s and vertical speed of ~5 m/s (Altas et al., 2020), the smallest resolved wavelength is 140 m. Hence, within the 10s moving window, the ~50 m in the integral vertical range is able to resolve the eddies smaller than 140 m, and preserve the potential of capturing the inertial subrange.

The use of TKE provides an illustration that in-cloud turbulence during ACE-ENA might be slightly stronger than that observed during SOCRATES. That being said, the quantitative evaluation of the turbulent enhancement of collision-coalescence requires access to the eddy dissipation rate, as typically used in model parameterizations (Grabowski and Wang, 2013; Wittle et al., 2019). The smallest scales resolvable with the 1Hz measurement used in this study are on the order of 140 meters, thus capturing only the larger-scale end of the inertial subrange and larger turbulent motions. Consequently, the ability to resolve smaller eddies and turbulent structures, crucial for understanding the energy cascade within the inertial subrange, is limited by the too-coarse spatial and temporal resolutions and aliasing issues (Siebert et al., 2010; Muñoz-Esparza et al., 2018; Kim et al., 2022). Therefore, to fully resolve the spectrum of turbulence and quantitatively examine energy dissipation and mixing processes, access to higher-frequency measurements is required to capture smaller eddies within the inertial subrange (Siebert et al., 2010; Lu et al., 2011; Waclawczyk et al., 2017). Additionally, the further quantification of the entrainment-mixing mechanisms also requires high-frequency eddy dissipation and accurate examination of the mixing time scale (Lehmann et al., 2009; Lu et al., 2011) for individual profile. Though currently beyond the scope of this study, those mechanisms will be of interest for future investigations.’

I will note that your point that the activation fraction of available CCN to cloud droplets is highly correlated with turbulence intensity (as stated in the abstract, albeit differently worded) is valid, but this is not quantitatively demonstrated either; you could adopt a framework as in Hu et al. (2021, doi: 10.1029/2021JD035180) to explore this point further.

Following the notion in Hu et al. (2021), we have calculated the correlation coefficients between the CCN activation ratio and the TKE, as an inter-cloud assessment. The following discussions are added to Section 4.1:

‘Previous studies have shown that the enhanced vertical turbulence (updraft velocity) can effectively facilitate CCN replenishment into the cloud layer (Hu et al., 2021; Zheng et al., 2022a&b) and increase the actual in-cloud supersaturation (Brunke et al., 2022), thus leading to a more efficient cloud droplet formation, enhancing the $ACI_{N,CB}$. By correlating the mean TKE values with the CCN activation ratio ($N_c/N_{CCN,0.35\%}$) for all individual cloud cases, the three IOPs show moderate but statistically significant correlation coefficients of 0.36, 0.55, and 0.51 for ACE-ENA summer, winter, and SOCRATES, respectively. This result reinforces the notion that the CCN activation fractions, particularly during the wintertime ACE-ENA, are significantly correlated with in-cloud turbulence intensities.’

And in the summary section:

‘The moderate but statistically significant correlation coefficients between the CCN activation fractions and the TKE agree with a previous study that found the local activation fraction of CCN to be strongly associated with increased updrafts (Hu et al., 2021).’

3. Beyond the lack of quantitative evidence supporting the hypothesis that turbulence is the cause of increased drizzle production during ACE-ENA winter, **there is a simpler, more parsimonious explanation that I believe is given short shrift in the manuscript: that a combination of a deep cloud layer along with relatively clean aerosol conditions during ACE-ENA winter results in robust drizzle generation**. You do discuss the relationship between cloud depth and precipitation susceptibility very briefly in section 4.2, but it essentially reads as a footnote versus a primary point. Given the strong dependence of cloud base rain rate on cloud depth in the empirical relation discussed in this same section (4.2), it was a major oversight that you didn’t explore this aspect further.

We agree with your argument and have revised and included the following discussion on the end of Section 3.3:

‘Drizzle formation and evolution in the ACE-ENA winter clouds are noticeably stronger than in the other two IOPs, which could be attributed to multiple factors. First, the ambient aerosols and CCN during winter are substantially fewer, featuring clean environments that promote the formation of generally larger cloud droplets due to the availability of more water content per droplet. Larger cloud droplets are more likely to collide and coalesce into drizzle drops, leading to relatively heavier precipitation (Chen et al., 2011; Duong et al., 2011; Mann et al., 2014). Furthermore, the wintertime clouds feature deeper cloud layers with mean thickness of (392.4 m) compared to the summertime clouds (336.3). In a thicker cloud layer with sufficient turbulence, the residence times of large cloud droplets and drizzle drops are elongated, and the chance of collision-coalescence growth can be effectively increased by recirculating the drizzle drops (Brost et al., 1982; Feingold et al., 1996; Magaritz et al., 2009; Ghate et al., 2021). Additionally, the prevalence of precipitation-evaporation-induced MBL cold pools, which disturb the MBL thermodynamics and contribute to turbulent mixing (Zuidema et al., 2017), during the wintertime might provide strong dynamical forcing to the warm-rain process (Jenson et al., 2021; J. Wang et al., 2022). As a result, the ACE-ENA wintertime drizzle DSD is sufficiently broadened, and the D_{mmd} is enlarged toward the cloud base. In comparison, although the SOCRATES exhibits even thicker clouds (487.4 m), the drizzle processes are seemingly suppressed by the much higher ambient aerosol and CCN concentrations.’

4. The discussion of the role of thermodynamic decoupling is incomplete, but decoupling ostensibly plays a significant role in the low values of fad encountered during all three campaigns. It would be well worth taking the next step and directly quantifying at least one decoupling metric from the observed profiles as defined by Jones et al. (2011).

We adapted the notion of Jones et al. (2011) and calculate the coupled layer and the degree of decoupling for every individual profile, as described in the following discussion in last paragraph of Section 2.1:

‘Jones et al. (2011) suggested that the MBL would be in a well-mixed and coupled condition when the difference in liquid water potential temperature (θ_L) and total water mixing ratio (q_t) between the bottom of MBL and the inversion layer are less than 0.5 K and 0.5 g/kg, respectively. In this regard, since the coupled and decoupled MBL conditions coexist in the selected cloud cases in this study, particularly in ACE-ENA summer, which is characterized by anomalously low BL heights and substantial BL decoupling. Previous studies found that, under the decoupling condition, the aerosols, CCN, and moisture sources near the surface are disconnected from the cloud layer aloft, hence exerting much less effective impact on the cloud microphysics (Zheng et al., 2022a; Christensen et al., 2023). Therefore, we adapt and modify the metric in Jones et al. (2011) to calculate the sub-cloud coupled layer, in order to ensure the aerosols and CCN measured sub-cloud are in a well-mixed state and can represent the actual interaction (or contact) with the cloud layer. In this study, the q_t and θ_L at the cloud base are calculated, and then their vertical variations are examined starting from the altitude of cloud base (z_b) and looking downward. As such, the coupled point altitude (z_{cp}) is defined as the altitude where the vertical changes in q_t and θ_L exceed 0.5 K and 0.5 g/kg, respectively. Hence, the coupled layer ($H_{cp} = z_t - z_{cp}$) is defined as the layer between the cloud top altitude (z_t) and coupled point altitude (z_{cp}), hence the selection of the aerosols and CCN within the below-cloud part of the coupled layer can be viewed as in contact with the cloud. An example of the coupled layer identification is shown in Figure S2. Therefore, the degree of MBL decoupling (D_{cp}) can be quantified as the ratio of the coupled sub-cloud MBL thickness to the sub-cloud MBL thickness, where $D_{cp} = 1 - (H_{cp} - H_c)/z_b$. As shown in Table S1, the ACE-ENA summer feature with highest degree of decoupling (averaged D_{cp}=0.504), compared to the ACE-ENA winter (D_{cp}=0.370) and SOCRATES (D_{cp}=0.277).'

5. I am not familiar with "retrospecting" as discussed in section 4.3 and shown in Fig. 8. What is the procedure for performing this analysis? Please explain in the manuscript, as this appears to be simultaneously one of the most tentative aspects of the paper as well as something the authors are rather excited about.

We have added the following discussion on the methodology:

'In order to examine the potential impact of the aforementioned processes on the ACI assessment, a sensitivity analysis is conducted by simply retrospecting the sub-cloud $N_{CCN0.35\%}$ according to their L_{CCN}. For each retrospective time step ΔT, the r_c values are held unchanged, and the retrospective $N_{CCN0.35\%}$ values for individual cloud cases are given by $N_{CCN0.35\%} - L_{CCN} * \Delta T$, and then the $ACI_{r,CB}$ can be recalculated. Note that assuming a constant r_c value over time inevitably induces uncertainty and biases, as it does not consider the microphysical processes affecting the cloud droplet mean size. However, previous numerical experiments show that the noticeable impact on the cloud mean radius through collision-coalescence necessitates a high degree of CCN depletion, and the quantified percentage changes in droplet mean sizes are several times less than the changes in CCN depletion (Feingold et al., 1996). Hence, the retrospective method, from an observational snapshot point of view, provides a direction that enables the assessment of $ACI_{r,CB}$ as if before the sub-cloud aerosols and CCN are scavenged by in-cloud coalescence-scavenging and precipitation scavenging processes.'

SPECIFIC COMMENTS

Each comment refers to a specific line/passage, figure, table or caption. Specific line(s) are denoted by LXX (or LXX-YY for longer passages).

L24-26: the lack of sensitivity of precipitation to aerosol during SOCRATES suggests that it inhabits a different microphysical regime than ACE-ENA. In other words, there are sufficient CCN during SOCRATES that the ACPI are effectively "saturated" with respect to increasing aerosol loading. There are numerous references discussing such buffering effects (a good starting point is Stevens and Feingold 2009, doi: 10.1038/nature08281) that I recommend the authors consult to reframe this discussion.

We have added the following discussion in Section 4.2 when discussing the precipitation susceptibility:

'…the ACE-ENA winter feature with enhanced collision-coalescence and the drizzle-recirculating processes, especially under low N_c conditions with more larger drizzle drops, leading to the increasing the S_o values. In comparison, the higher ambient aerosol and CCN concentrations during SOCRATES lead to relatively narrower drizzle DSDs and may induce effective aerosol buffering effects, where the warm-rain processes in cloud are already fairly suppressed, hence diminishing the sensitivity of R_{CB} to N_c (Stevens and Feingold, 2009; Fan et al., 2020; Gupta et al., 2022).'

L154-157: with respect to what are the "changes" in θ_L and qt evaluated? The mean of the cloud layer or surface layer? Based on Fig. S1, I assume cloud layer. I am confused by this definition because "mixed layer" typically implies surface mixed layer, while you are using it to describe an elevated mixed layer. In general, I found your analysis of decoupling to be incomplete.

We have refined our discussion, please refer to our previous response on the MBL decoupling discussion.

L191-195: When you discuss “airmass origin,” at what vertical level(s) are back trajectories taken? I believe you in terms of PBL airmass, but is this also true above the PBL?

We have expanded the statements on the airmass origin as follows:

‘In the SOCRATES region, according to the previous studies involving back-trajectory analyses, dominant air masses within the MBL primarily originate from the south or from the west, skirting the Antarctic coast (Zhang et al., 2023), while the air masses above the MBL follow a similar transport pathway, they can also originate from the tip of southern Africa and transport southeast along the warm conveyor belt (McCoy et al., 2021).

Conversely, the ENA region experiences aerosols of varied origins, spanning maritime air masses to those heavily influenced by continental emissions from North America or Northern Europe, especially during the summertime (Logan et al., 2014; Wang et al., 2020). The summertime air mass back-trajectories within the MBL strongly feature recirculating flow around the Azores high. During the wintertime, however, the air masses predominantly originate in the FT, are transported above the MBL, and are then further entrained down to the MBL by large-scale subsidence, indicating less influence from continental pollution (Y. Wang et al., 2021b).’

L192 and elsewhere: Zhang et al. (2023) reference is missing in bibliography – I assume the correct reference is from (most of) the same authors in Atmosphere (doi: 10.3390/atmos14081246)?

Yes, we have added the correct reference to the reference list. Thanks for catching.

L216-217: please add a location for the further discussion, i.e., “and will be further discussed in Section X.X” or “further discussed later in this section”

We have added the location for further discussion as: '…and will be further discussed in Section 3.1.'

L259: are there any measurements that support your assertion that it's both coalescence-enlargement and sea salt contributing to heightened concentration at Dp>1 um?

We have added the reference on previous case analysis during the summer ACE-ENA (Zheng et al., 2022b), and the long-term statistics on the coarse-mode aerosol seasonal variation over the ARM-ENA site (Zheng et al., 2018) to back up this statement.

'The elevation in sub-cloud coarse mode aerosols observed for both ACE-ENA IOPs (as seen in Fig. 2) can be attributed both to the coalescence-enlargement process and the intrusion of sea spray aerosols (e.g., sea salt). As illustrated and analyzed based on a case study during summertime that exhibits the signal of cloud-processing aerosols (Zheng et al., 2022b), as well as the long-term aerosol physicochemical properties over the ARM-ENA ground-based observatory (Zheng et al., 2018), particularly during the winter season where the production of sea spray aerosol is prevalent.'

L268-269: I don't think you've improved understanding of the first indirect effect. Rather, you're adding another data point that supports what we already understand about it. So it's more of a "confirmation" than anything novel.

We have eased the tone and changed this statement to: 'These results have further confirmed and reassured our understanding of the aerosol first indirect effect.'

L296: Verb tense disagreement. Recommend you make everything present tense: "…air **is** entrained into the boundary layer…"

We have changed this statement to present tense as 'The warmer and drier air near the cloud top entrains into the cloud layer and further mixes downward, often resulting in the evaporation of small cloud droplets and the shrinking of droplet sizes, which oppose condensational growth (Desai et al., 2021).'

L300-302: Does this difference in rc profiles tell you anything about mixing regime (i.e. homogeneous vs. inhomogeneous)?

We have added the following discussion on the entrainment mixing regime:

'For the three IOPs, the N_c and LWC_c exhibited a stable trend from the cloud base, followed by a noticeable decrease near the cloud top mixing zone, while the changes in r_c trend were not as dramatic as the others. Such characteristics of the cloud microphysics vertical profiles indicate the signal of inhomogeneous mixing, which occurs when dry and warm air mixes unevenly and not rapidly with the cloud air, hence partially evaporating the cloud droplets (Lehmann et al., 2009; Lu et al., 2011). The results are consistent with findings in stratocumulus clouds over multiple field campaigns (Brenguier et al., 2011; Jia et al., 2019) and with the findings for selected cases during the ACE-ENA (Yeom et al., 2021), and the SOCRATES (Sanchez et al., 2020). While the near-cloud r_c profiles for the ACE-ENA cases exhibit more constant variation, which could be possibly attributed to more effective mixing due to the stronger entrainment rate, particularly during the ACE-ENA winter, eventually reaching a smaller equilibrium in terms of mean sizes.'

L304: I can't imagine the water vapor source from entrainment evaporation is a leading order term in the BL qt budget, and I'm having some difficulty understanding the relevance of raising this point. As you state, the net impact of entrainment on BL qt should be negative (i.e., entrainment mixes in drier air, so BL-mean qt should decrease), which would imply this evaporative source is a relatively minor offset to the entrainment drying sink. Eyeballing it from Fig. 3c, it looks like there's maybe 0.2 g/m3 of vapor that is liberated from the clouds (extrapolating the midcloud ql lapse rate to zi=1) – but I can't assess this any further since you don't show any mean qt or qv profiles. Both the G1 and the GV have open path hygrometers from which qv can be accurately measured in cloud – if you want to get into a discussion of the vapor budget, it would be helpful to explicitly show some of these measurements.

We are not intended to suggest that the evaporation will cause a difference in an order of magnitude. We appreciate you consideration on this argument, however, we have considered to delete this statement to avoid further confusion, and changed it to:

‘As cloud-top entrainment mixing can shrink large cloud droplets via evaporation, depending on the entrainment mixing rate, the nearly constant r_c values (at $z_i > 0.8$) might represent the equilibrium balance between two competing processes: cloud droplet condensational and collision-coalescence growths, and the entrainment mixing evaporation effects.’

L305-310: What evidence do you have for re-condensation beyond the inferences made from bulk profiles? And shouldn’t there be *more rapid* growth on smaller drops since condensation rate is inversely proportional to surface area? You have the full DSDs to demonstrate the validity of the generalizations you’re drawing. Please evidence for these assertions.
L313: What is gained by quantifying Δr_C? Is this not just a different way of expressing the subadiabatic ql lapse rate via the relation $r_e = kr_v$ where $r_v \propto (q_l/N)^{1/3}$?

Response to L305-310 & L313:

We are trying to provide bulk descriptive discussion to introduce the discussion on the cloud microphysical responses on aerosols later, we have removed the argument on re-condensation and changed the discussion here as:

‘When dry air entrainment occurs at the cloud top, some of the upper-level smaller cloud droplets will evaporate, which leads to decreases in N_c (Fig. 3a). As cloud-top entrainment mixing can shrink large cloud droplets via evaporation, depending on the entrainment mixing rate, the nearly constant r_c values (at $z_i > 0.8$) might represent the equilibrium balance between two competing processes: cloud droplet condensational and collision-coalescence growths, and the entrainment mixing evaporation effects.

The increases of r_c (Δr_c) from cloud base to cloud top are 4.03 μm, 4.78 μm and 5.85 μm, with percentage increases of 66%, 68% and 79%, for SOCRATES, ACE-ENA summer and winter, respectively. Even though, theoretically, the condensational growth effect would be more pronounced on smaller cloud droplets due to their smaller surface area (Wallace and Hobbs, 2006), SOCRATES exhibits the thickest mean cloud thickness but experienced the least r_c increases among the three IOPs. This suggests that high aerosol loadings are limiting the overall growth of the cloud DSD in SOCRATES clouds, while the ACE-ENA winter clouds show the strongest r_c increase, in contrast. This comparison suggests different cloud microphysical responses to aerosol perturbations in the three IOPs, which will be further discussed in Section 4.1.'

L337-340: Please define what terms are being used to calculate the "reduction of LWC_C" – it's not clear to me what you're doing here.

We are comparing the LWC_c deficit between the three IOPs, from where it starts to decrease according to the mean profiles. We have added more description to the discussion:

'Considering the near cloud-top proportion of cloud where the LWC_c experienced decrease, the difference in LWC_c (between the cloud top value the upper-middle cloud maximum for the mean profiles) for the ACE-ENA summer (-0.032 g m^{-3}) is higher than the reductions in winter (-0.018 g m^{-3}) and SOCRATES (-0.009 g m^{-3}).'

L368-369: Water vapor competition matters in a water-vapor limited regime (which seems quite obvious when stated that way…), but it seems to me that ACE-ENA winter is more of an aerosol limited regime. Appealing to water vapor competition is not a "one size fits all" conclusion that can be universally applied.

We have added the following discussion in this regard:

'In addition, the discrepancies in ε between the three IOPs may be attributed to the sub-cloud aerosol differences, which essentially resided in different microphysical regimes. Y. Wang et al. (2021a) stated that higher aerosol loading would lead to increased ε due to the water vapor competition effect, supporting the discrepancy between SOCRATES and ACE-ENA summer IOPs, which can be categorized as a water-vapor-limited regime. Meanwhile, the ACE-ENA wintertime IOP exhibits characteristics of an aerosol-limited regime, in which the cloud DSDs tend to be narrower than in the water-limited regime, due to enhanced droplet growth, and the ε values further decrease with height via the condensational narrowing effect (J. Chen et al., 2018).'

L370: do you quantify skewness or is this a qualitative description?

We have calculated the skewness values for the cloud DSDs and include them in the discussion: 'For the four cloud portions from cloud base to cloud top, the skewness of summertime (wintertime) cloud DSDs are 0.627 (0.271), 0.358 (0.175), 0.098 (-0.063), and -0.362 (-0.554), respectively.'

L371-372: can you say with certainty whether coarse mode aerosols are drizzle residuals vs. “primary production” of sea salt from the surface? Seems like a difficult “chicken and egg” problem to assess from in situ data without either aerosol composition information or some modeling work to back up the statement.

Since both campaigns lack continuous coarse mode chemical compositions and the offline analyzed samples are inadequate for the select cases. We have added the following discussion regarding this issue:

‘These coarse mode aerosols, whether from primary production of sea spray or from the residuals of evaporated drizzle drops, are more easily activated (or re-activated) into larger cloud droplets when they intrude (or recirculate) into the cloud layer (Hudson and Noble, 2020; Hoffmann and Feingold, 2023). Nevertheless, it is challenging to pinpoint the actual origins of coarse mode aerosols from the perspective of aircraft observational snapshots, thus requiring further numerical modeling work.’

L436: I assume you mean liquid water *content*, but this is not stated

We have changed this statement to:

‘Furthermore, the similarity in the vertical integral of LWC_c (as shown in Fig. 3c) provides comparable liquid water between three IOPs’

L490-491: What are the uncertainties in $S0$? The correlations do not look very strong in Fig. 7a.

The S_0 values are 0.979, 1.229 and 1.638, with the absolute values of correlation coefficients are 0.33, 0.29 and 0.45, for SOCRATES, ACE-ENA summer and winter, respectively. These correlation coefficient values fall within the reasonable range found in previous studies on precipitation susceptibility in MBL stratus and stratocumulus clouds (Jung et al., 2016; Gupta et al., 2022), and indicate statistically significant (but not strong) dependences of R_{CB} on N_c, since the R_{CB} is not solely dependent on the N_c.

L514: Double check the equation, it looks like something is not formatted correctly or there are some extra characters.

We have corrected the equation:

$$R_{CB} = 1.73e^{-10}\ H_c^{3.6} N_a^{-1}$$

L536: Should there be a minus sign in the 2nd parenthetical?

Yes, we have corrected it.

L538-539: This sentence needs to be restructured, it is currently a fragment.

We have changed it to 'As the results indicate, the ACE-ENA clouds experience more substantial sub-cloud CCN loss than SOCRATES, especially in wintertime precipitating clouds.'

L542-543: you could expand upon this point more, it's a bit too concisely expressed to be easily understood.

We have further expanded on this discussion as:

'Recall that the assessment of $ACI_{r,CB}$ relies on the relative changes of r_c and N_{CCN}, while the different L_{CCN} for individual cases can result in the shrinking of the N_{CCN} variation ranges (imagine the abundant CCN are depleted by the coalescence-scavenging). In other words, the given change in r_c corresponds to a narrowed change in N_{CCN}. Mathematically speaking, the assessment of $ACI_{r,CB}$ depends on the ratio of the numerator (change in r_c) and the denominator (change in N_{CCN}). Under the circumstances of substantial cloud-processing to the aerosols, the altered sub-cloud CCN budgets are reflected as a smaller denominator, versus the less altered numerator, hence mathematically presented as an enlarged $ACI_{r,CB}$.'

L569: it is rather counterintuitive that the "pristine" environment has the strongest aerosol loading. This is paradoxical because we often use the terms "pristine" vs "polluted" to imply low vs. high aerosol loading, respectively. You clearly mean it in the sense that the SO region is minimally impacted by anthropogenic emissions. So a little word-smithing is needed to resolve this incongruity.

We have changed the term ‘pristine’ to ‘pre-industrial’ in order to describe the SOCRATES nature, as well as in the relevant discussion throughout the revised manuscript.
E.g., ‘The SOCRATES features the pre-industrial natural environment enriched by aerosols from marine biological productivity and without the contamination of anthropogenic aerosols.’

L594: fully agreed that the assumption of constant fad used in satellite N retrievals is problematic, but how do the campaign-average profiles presented here improve this situation? On a profile-by-profile (or, from the satellite perspective, pixel-by-pixel) basis, is there anything from the measurements that suggest potential predictors of fad? Or do you view your contribution as simply another data point showing that an assumed value of 0.8 is unrealistic?

We have eased our tone and modified our discussion to:

‘While satellite retrievals of droplet number concentration heavily rely on the adiabatic cloud assumption and are usually given as a constant of f_{ad} = 0.8, the in-situ observational evidence found in this study further confirms the unrealistic nature of this assumption. It will be of interest to utilize multiple aircraft measurements (campaigns) to explore the variability of MBL cloud and drizzle microphysical properties over different marine regions. This can help examine potential predictors for f_{ad}, which will aid in satellite-based retrievals and aerosol-cloud interaction assessments (Painemal and Zuidema, 2011; Grosvenor et al., 2018; Painemal et al., 2021).’

Figure 2: This figure could be compressed in the horizontal, which would accentuate the shape of the distribution in a manner pleasing to the eye. As it currently stands, this looks “stretched out” and there aren’t many interesting detail/wiggles in any of the curves that merit such a long aspect ratio.

Figure 2 is replotted with smaller aspect ratio.

Figure 3: What do the shaded regions denote? Interquartile range? Standard deviation? 5th-95th percentiles? No info in caption.

The shaded region denotes the inter-cloud-case standard deviation. We have added this information to the captions.

Figure S2: put the two panels on the same plot so they can be directly compared.

Figure S2 is updated accordingly.

Figure S3: why does this size range needs to be separated from Fig. 2 in the main manuscript?

The coarse-mode range is added to the new Fig. 2, and the original Fig. S3 is removed.

Figure S4: please add uncertainty shading as you did in Fig. 3 of the manuscript, it would be helpful to see the variability of subadiabaticity within campaigns
The standard deviations are added as shading areas in Fig. S4.

Table S1: Please include f_ad in this table.

The f_{ad} values are included.

Table S2: it looks like this table is cutoff. Are there more variables not shown?

We have fixed the table appearance.

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