Response to Reviewer #2

We appreciate your time for carefully reviewing our manuscript. We would like to thank you for the constructive comments and suggestions, which encourage and help us to improve the manuscript. The manuscript has been revised accordingly. In the response below, your comments are provided in black text and our responses are provided in blue text.

Response:

As the title suggests, this paper describes an aggregated analysis of aerosol-cloud interaction (ACI) in non-precipitating marine boundary layer clouds at the Eastern North Atlantic ARM remote sensing supersite. A relatively narrow view of ACI is taken in which the bivariate relationship between aerosol and cloud drop number concentration and the ACI index were calculated numerous times, compositing by various column-mean or column-integral quantities (e.g., water vapor path, cloud adiabaticity, lower tropospheric, turbulence). My main concern with the study is that each of these purported controlling factors is analyzed in isolation, which implicitly assumes no covariability among them. This assumption is not valid and no attempt to address this issue was given. As such, I find it difficult to accept many of the mechanistic arguments made by the authors. They cannot demonstrate cause and effect, and there are clearly confounding variables that limit their ability to draw stronger conclusions (for example, lines 243-244: "the coincidence of high NCCN and PWV does not necessarily imply a physical relationship"). I therefore recommend the manuscript be rejected and the authors encouraged to resubmit after broadening their analysis. The premise of evaluating ACI with the authors' retrieval product is promising, but to understand the role of the controlling factors, they must be analyzed in a multi-dimensional framework (principal component analysis, k-means clustering, etc.) that allows the authors to identify and, more importantly, interpret co-variability among environmental factors. As it currently stands, the conclusions of this study point vaguely toward correlations with large-scale variables but give no clear guidance.

Thanks for the constructive suggestions. To better address the reviewer's concern about the co-variabilities between the environmental variables and to more clearly shed light on their impacts on ACI, we have now conducted the principal component analysis (PCA). The variables of sub-cloud precipitable water vapor (PWV_{BL}), the boundary layer decoupling index (D_i), the vertical component of the turbulence kinetic energy (TKE_w), the lower tropospheric stability (LTS) and the surface wind directions in terms of northerly and southerly ($W_{dir,NS}$) are constructed as the input of the eigenanalysis. Results show that the first three PCs can describe the majority (~84%) of the variance among the selected variables. Where the most explanatory PC1 (account for 43.72% contribution) strongly correlated with PWV_{BL}, D_i (both negatively) and TKE_w (positively), and hence describe the co-variation of the boundary layer conditions. While the PC2 and PC3 (account for 22.01% and 18.26% contributions, respectively) are strongly correlated with the LTS and $W_{dir,NS}$, which likely indicates the variations of the Azores High position and strength. By projecting the variables onto PC1 and PC2, the PCA loading analysis shows that the TKE_w are strongly negatively correlated with D_i, which as expected since a more decoupled MBL is often separated into two layers where the lower one can cap the surface moisture, while the higher TKE_w denote sufficient turbulence that maintains the well-mixed MBL. Additionally, the island effect is also indicated by the eigenanalysis, where the surface northerly wind would induce additional updraft velocity and hence disturb the TKE_w, owing to the topographic effect of the cliff north of the ENA site. Upon the PCA results, the role of cloud adiabaticities on the behaviors of CCN-N_c conversion is further examined using both binning and eigenanalysis. And the factors that have the most influence on the explanatory PCs are selected as the sorting variables in the ACI_r assessments.

The detailed discussions on the multi-dimensional PCA have been added to the section 3.4 of the revised manuscript as follows:

3.4 The co-variabilities of the meteorological factors

The environmental conditions over the ENA have been widely studied as not independent but entangled with each other (Wood et al., 2015; Zheng et al., 2016; Wu et al., 2017; Wang et al., 2021). To better understand the dependencies and the co-variabilities of the meteorological factors, a principal component analysis (PCA) is performed targeting on the following variables: (1) PWV_{BL} denotes the water vapor availabilities within the boundary layer; (2) D_i describes the boundary layer coupling conditions; (3) TKE_w represents the strength of boundary layer turbulence; (4) $W_{dir,NS}$ reflects the surface wind directions in terms of northerly and southerly; and (5) LTS infers the large-scale thermodynamic structures. Note that the $W_{dir,NS}$ are taken as $W_{dir,NS} = abs(W_{dir} - 180°)$, so that the original W_{dir} (0-360°) can be transformed to $W_{dir,NS}$ (0-180°) where the values smaller than 90° are close to the southerly wind, and those greater than 90° are close to the northerly wind. The $W_{dir,ns}$ are transformed as such to capture the island effects better, because the cliff is located north of the ENA site.

The input data metric is constructed from the above five variables to apply the PCA, and the principal components (PCs) that serving to explain the variation of those dependent variables can be output from the eigenanalysis. The result shows that for the five selected meteorological factors, the proportions of the total intervariable variance explained by the PCs are 43.72%, 22.01%, 18.26%, 8.95% and 7.06%, and the eigenvalues are 2.19, 1.10, 0.91, 0.45, and 0.35, respectively. Note that the first three PCs have the highest eigenvalues and explain most (~84%) of the total variance, which indicates that they can capture the significant variation patterns of the selective meteorological factors.

To determine the relative contributions of the variables to PCs, all the five selected meteorological variables are projected to the first three PCs and the Pearson correlation coefficients between them are listed in Table 4. For the first PC (PC1) which accounts for the highest proportion (43.72%) of the total variance, the PC1 is strongly negatively correlated with PWV_{BL} (-0.84) and D_i (-0.73), but strongly positively correlated with TKE_w (0.69). These results suggest that PC1 mainly represents the boundary layer conditions, and the co-variations of the boundary layer water vapor and turbulence are the most distinct environmental patterns for the selected cloud cases. The PC2 and PC3 are most correlated with LTS (0.58 and 0.65 for PC2 and PC3, respectively) and $W_{dir,NS}$ (0.60 and -0.50 for PC2 and PC3, respectively), indicating that the PC2 and PC3 mainly describe the variations in large-scale thermodynamic and the surface wind patterns, which are likely associated with the variations of the Azores High position and strength (Wood et al., 2015).

Table 4. The first three principal components from eigenanalysis

Eigenanalysis	PC1	PC2	PC3
Eigenvalues	2.17	1.10	0.91
Proportion of variance explained (%)	43.72	22.01	18.26
Cumulative proportion (%)	43.72	65.73	83.99
Correlations (Variables vs. PCs)	**PC1**	**PC2**	**PC3**
PWV_{BL}	-0.84	0.20	-0.11
D_i	-0.73	-0.48	-0.20
TKE_W	0.69	0.35	-0.44
$W_{dir,ns}$	0.52	0.60	-0.50
LTS	-0.43	0.58	0.65

To further understand the correlations between the meteorological variables, the principal component loadings plot is constructed by projecting the variables onto PC1 and PC2 as shown in Fig. 4. Each point denotes the variable correlations with PC1 (x-coordinate) and PC2 (y-coordinate), so that each vector represents the strength and direction of the original variable influences on the pair of PCs. The angle between the two vectors represents the correlation between each other. In Fig. 4, both $\mathrm{TKE_w}$ and $W_{dir,NS}$ vectors are located in the same quadrant (positive in both PC1 and PC2) and close to each other with a small degree of an acute angle, which means the $\mathrm{TKE_w}$ are strongly correlated with the $W_{dir,NS}$. When the surface wind is coming from the north side of the island, the topographic lifting effect of the cliff would induce additional updraft over the ENA site (Zheng et al., 2016), so that the wind closer to the northerly wind (larger $W_{dir,NS}$) is more correlated with higher $\mathrm{TKE_w}$. Note that $\mathrm{TKE_w}$ and D_i vectors are almost in an opposite direction, which denotes a strongly negative correlation between the two variables. The angles of $\mathrm{PWV_{BL}}$ with D_i (~45°) and $\mathrm{TKE_w}$ (~142°) suggest that $\mathrm{PWV_{BL}}$ is moderately positively correlated with D_i but negatively correlated with $\mathrm{TKE_w}$. A higher D_i indicates a more decoupled MBL, where MBL is not well-mixed and separated into a radiative-driven layer and a surface flux driven layer that caps the surface moisture (Jones et al., 2011). This situation is more likely to associate with a relatively higher $\mathrm{PWV_{BL}}$ and weaker $\mathrm{TKE_w}$ condition. As for the LTS parameter, the close to 90° angle with $\mathrm{TKE_w}$ suggests no correlation between them, since the LTS is mostly capturing the large-scale thermodynamical structures and is obtained from a coarser temporal resolution. Thus, the LTS does not essentially have correspondence to the strength of boundary layer turbulence and can be treated as independent to $\mathrm{TKE_w}$ over the ENA site. The loading plot intuitively tells us the directions and strengths of the co-variabilities of the selected meteorological variables, and sheds the light on determining the key factors that are feasible to use in examining the environmental impacts on the aerosol-cloud interactions.

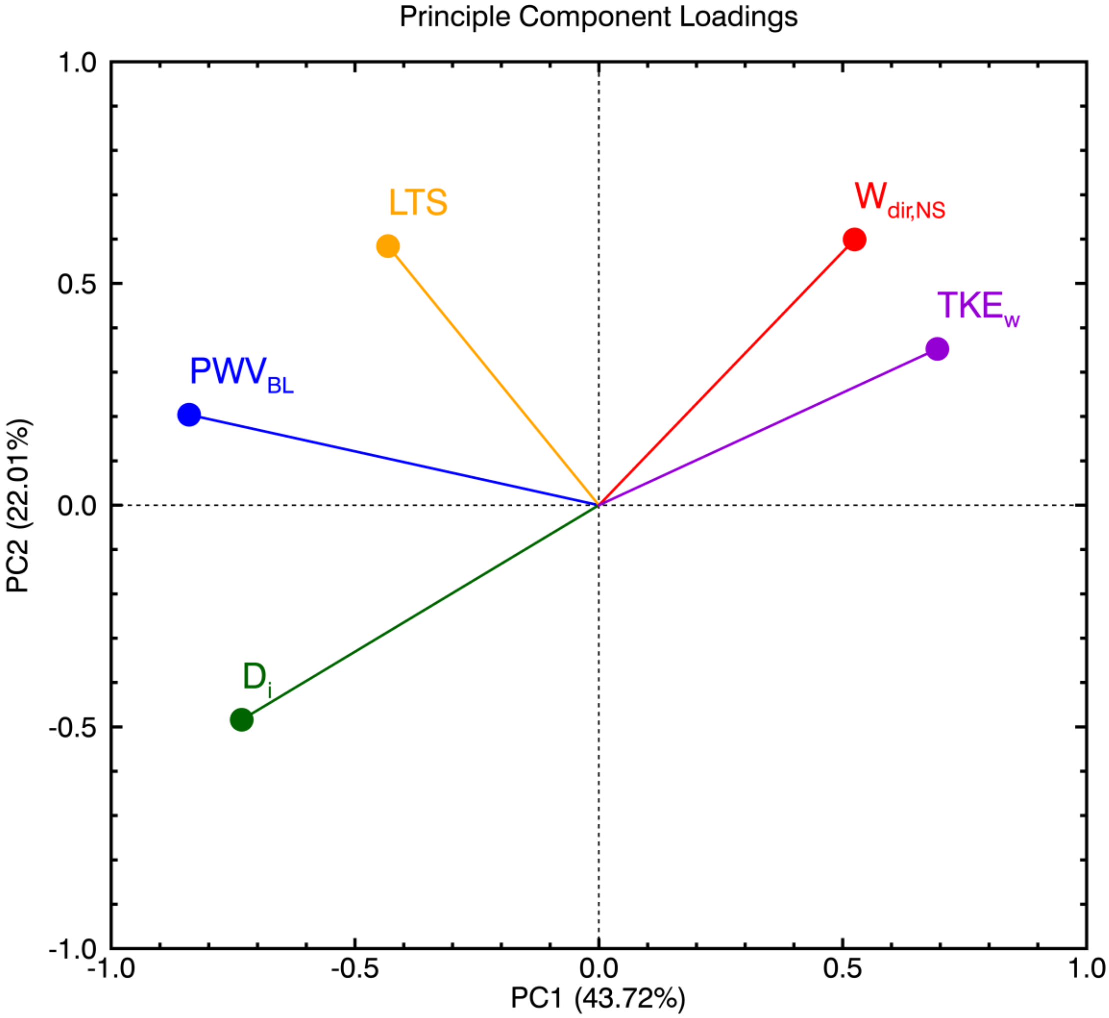

Figure 4. The projections of TKE_w (purple), $W_{dir,NS}$ (red), LTS (orange), PWV_{BL} (blue) and D_i (green) onto the first principal component (PC1) and the second principal component (PC2). The x-coordinates denote variables' correlations with PC1, and the y-coordinates denote variables' correlations with PC2.

In addition, the detailed results and discussions on the impacts of meteorological factors on aerosol and cloud properties, and aerosol-cloud interactions can be found in the section 3.5 of the revised manuscript.

I have a number of other concerns the authors may also wish to consider:

- How good of a proxy is PWV for PBL relative humidity? Are there cases when non-drizzling stratocumulus occur with a relatively moister free troposphere? Perhaps you could estimate the fraction of PWV in the PBL using the interpolated sonde product or Raman lidar (note: Raman will only get you subcloud vapor)?

Thanks for the comment and suggestions. In the revise manuscript, we changed to use the sub-cloud boundary-layer PWV (PWV_{BL}), and tested the contribution of PWV_{BL} to column PWV. The discussion has been added to the section 2.2 in the revised manuscript as follows:

To capture the information of MBL water vapor more accurately, the sub-cloud boundary layer integrated precipitable water vapor (PWV_{BL}) is calculated using the interpolated sounding product following:

$$PWV_{BL} = \frac{1}{\rho_w} \sum (z_{i+1} - z_i) * (\rho_{v,i+1} + \rho_{v,i})/2, \quad (1)$$

where the ρ_w is the liquid water density and the ρ_v is the water vapor density collected from the Interpolated Sounding and Gridded Sounding Value-Added Products (Toto and Jensen, 2016), the subscripts i and $i+1$ represent the bottom and top of each interpolated sounding height layer. Both PWV and PWV_{BL} are temporally collocated to 5-min resolution and plotted against each other in Fig. S1a to test the contribution of PWV_{BL} to the PWV. The Pearson correlation coefficient of 0.85 shows that the PWV_{BL} are strongly positively correlated with the PWV, while the distribution of the percentage ratio of PWV_{BL} to PWV (Fig. S1b) indicates that, on average, the PWV_{BL} contribute to ~58% of the PWV. Considering the cloud-topped MBL, the majority of cases (~74%) associate with a relatively moist boundary layer compared to the amount of water vapor in the free troposphere, where the PWV_{BL} already contributed over 50% of the total column PWV. In contrast, only ~9% of cloud samples occur under a relatively dry boundary layer and moist free troposphere, where PWV_{BL} contributions are less than 40%. In general, the PWV can well capture the variation of the PWV_{BL}. In the rest of the study, the PWV_{BL} are used, as it represents the sub-cloud boundary layer water vapor availabilities which are more closely related to the MBL cloud processes.

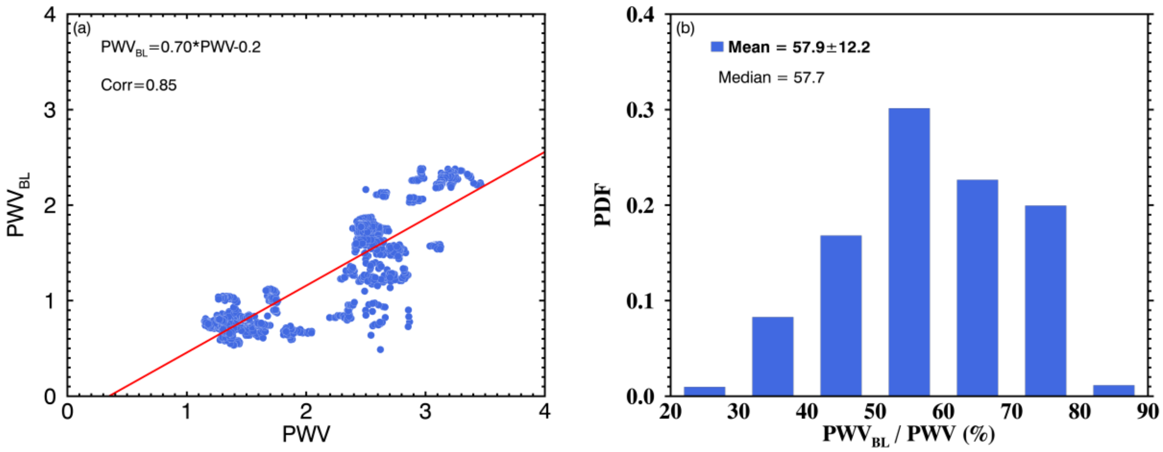

Figure S1. (a) Scatterplot of PWV versus PWV_{BL}; and (b) distribution of the percentage ratio of PWV_{BL}/PWV.

- Not enough information is given about how the vertical velocity variance TKEw is calculated. Is it a PBL average? A Doppler lidar column-deep average? Column max. value? And what Doppler lidar product are you using to get variance? The standard 10-minute integration? The median value seems low for surface-coupled stratocumulus cases. Are you evaluating any decoupled cases? There is also a diurnal and season cycle of turbulence at this site (at least, when sampling an undisturbed marine airmass; see more below), which may also be affecting your statistics.

In this study, the vertical component of the turbulence kinetic energy (TKE_w) are used, which is defined as:

$$TKE_w = \frac{1}{2}\overline{(w')^2} , \quad (2)$$

where the $(w')^2$ is the variance of vertical velocity measured from the Doppler lidar standard 10-min integration, which collected in the Doppler Lidar Vertical Velocity Statistics Value-Added Product (Newson et al., 2019). The noise correction has been applied to reduce the uncertainty of the variance to ~10% (Hogan et al., 2009; Pearson et al., 2009). In this study, the mean value of TKE_w in the sub-cloud boundary layer proportion of the Doppler lidar range is used, and the data temporal resolution is further downscaled to 5-min for temporal collocation purposes.

The description of TKE_W above has been added to the section 2.2 of the revised manuscript.

We have also included the decoupling index (D_i) given by: $D_i = (z_b - z_{LCL})/z_b$, where the z_{LCL} is the lifting condensation level calculated analytically following the method in Romp (2017), with an uncertainty of around 5 m. The surface temperature, pressure, relative humidity, and mass fraction of water vapor that used in the z_{LCL} calculation, as long as the vector-averaged wind directions (in 360° coordinate) over the ENA site are obtained from the ARM surface meteorology systems (ARM MET handbook, 2011).

In this study, we are trying the examine the environmental effects on ACI_r under the diverse conditions and whether the ACI_r can be distinguished by them, so that we did not have prior selection on any particular environmental factors (except only the non-precipitating stratiform cloud cases), and thus the samples including strongly decoupled, moderate-to-loosely decoupled and coupled MBL conditions.

From the PCA, the TKE_w has been found to be strongly positively correlated with $W_{dir,NS}$ and negatively correlated with D_i, which means the values of TKE_w already account for the co-variabilities in these variables. Therefore, treating TKE_w as the sorting variables would lead to a more physical process-orientated assessment. And the corresponding discussion is revised in section 3.5.2 of the revised manuscript.

- Have you controlled for wind direction in your analysis? It has been shown that there is an island effect when the surface wind is from the island (e.g., Zheng, Rosenfeld and Li 2016). Overland flow affects boundary layer turbulence and may also impact surface fluxes, PBL depth and CCN composition.

We have considered the potential impact of the wind direction on the boundary layer turbulence, and added to the PCA. In addition, the following summary on the island effects has been added to section 3.4 of the revised manuscript:

In Fig. 4, both TKE_w and $W_{dir,NS}$ vectors are located in the same quadrant (positive in both PC1 and PC2) and close to each other with a small degree of an acute angle, which means the TKE_w are strongly correlated with the $W_{dir,NS}$. When the surface wind is coming from the north side of the island, the topographic lifting effect of the cliff would induce additional updraft over the ENA site (Zheng et al., 2016), so that the wind closer to the northerly wind (larger $W_{dir,NS}$) is more correlated with higher $\mathrm{TKE_w}$. Therefore, the values of $\mathrm{TKE_w}$ already account for the co-variation of $\mathrm{TKE_w}$ and $W_{dir,NS}$.

- How much does LTS tell us at a site like ENA, and what physical motivation do you have for including it as a sorting variable? I always envision LTS as having the most meaning in the subtropical eastern boundary current (EBC) areas, i.e., northeast/southeast Pacific and southeast Atlantic. The Azores are more of a mixed subtropical/midlatitude site that has much warmer SST than in the traditional EBC areas where MBL clouds are studied, and much of the cloud cover at ENA occurs in transient postfrontal subsidence vs. longer-lasting large-scale subsidence where the spatial gradient (of both subsidence and SST) matters more in defining cloud type transitions.

We agree with your comment that the LTS might not be a feasible variable to use over ENA site, we included the LTS as it is orthogonal to the $\mathrm{TKE_w}$ from the PCA and thus can be treated as independence. We have added the relative discussion in section 3.5.2 of the revised manuscript:

Combining LTS and $\mathrm{PWV_{BL}}$ as sorting variables, the $\mathrm{ACI_r}$ values for four regimes are shown in Fig. S4. The $\mathrm{ACI_r}$ differences between low and high $\mathrm{PWV_{BL}}$ regimes are still retained. In the low $\mathrm{PWV_{BL}}$ regime, the $\mathrm{ACI_r}$ values are limited to 0.016 and 0.056 for low and high LTS regimes, respectively. In the high $\mathrm{PWV_{BL}}$ regime, the $\mathrm{ACI_r}$ values are 0.150 and 0.171 for low and high LTS regimes, respectively, which is about 3-5 times greater than those in low $\mathrm{PWV_{BL}}$ regime. However, the $\mathrm{ACI_r}$ in different LTS regimes cannot be distinctly differentiated ($\mathrm{ACI_r}$ differences between LTS regimes are ~0.02 and ~0.04), and the main difference in $\mathrm{ACI_r}$ are still induced by the $\mathrm{PWV_{BL}}$. Owing to the location of the ENA site where it locates near the boundary of mid-latitude and subtropical climate regimes, the MBL clouds over the ENA are found to be often under the influences of cold fronts associated with mid-latitude cyclones, where the cloud evolutions are subject to the combine effects of post-frontal and large-scale subsidence (Wood et al., 2015; Zheng et al., 2020; Wang et al., 2021). Therefore, over the ENA, although the spatial gradient of LTS is studied to be associated with the production of MBL turbulence and the change in wind direction (Wu et al., 2017), the LTS value itself is examined to has a weak impact on the aerosol-cloud interaction from this study.

- For arguments you make about the relationship between entrainment, collision-coalescence and number concentration, it is problematic that your retrieval assumes constant Nc throughout the cloud layer. When entrainment-induced evaporation and/or collision-coalescence are active, this assumption is broken. In general, I don't understand your argument that entrainment is a sink of Nc.

The Wu et al. (2020a) retrieval works as separating the reflectivity to the contributions of cloud (Z_c) and drizzle, the cloud procedure assumes an initial guess of the representative layer-mean N_c based on the climatology over ENA sites (Dong et al., 2014), and such allows the first guess of the vertical profile of LWC based on N_c and Z_c, and then constrains back the N_c and LWC using the LWP from MWR, finally output r_e (Fig.3 in Wu et al., 2020a). Therefore, the final retrieved N_c is updated to in response to the cloud microphysical processes within this time-step. From the aircraft in-situ measurements during the ACE-ENA, we used the in-situ measurement during ACE-ENA to validate the retrieval outputs and found that the observed N_c profile is near-constant in middle part of the cloud, with the signal of entrainment-induced depletion near the cloud top, even in the drizzling cloud where the collision-coalescence processes are more active (Wu et al., 2020a). However, it is hard and beyond the scope of the ground-based retrieval to compare the vertical dependency of depletion rate within one time-step. Therefore, as the retrieval currently work as representing the layer-mean information from the given time-step, the preferred method in this study is to compare N_c at different times, where in this case are the adiabatic versus sub-adiabatic conditions which hence yields different N_c that we retrieved from the ground-based snapshot perspective. From the PCA and binning analysis, the effect of cloud adiabaticities on CCN-N_c conversions may shed light on interpreting the aerosol-cloud interaction under different environmental effects.

We have added the above discussion in section 3.5.1 of the revised manuscript.

- High CCN events at ENA are not only from North America. They have also been traced to North Africa and Europe.

The corresponding sentence is changed to 'A few instances of aerosol intrusions (~3%) with higher $N_{CCN,0.2\%}$ were likely a result of continental air mass transport from North America, Europe, and Africa (Logan et al., 2014; Wang et al., 2020).'

References.

Wu, P., Dong, X., Xi, B., Tian, J. and Ward, D. M.: Profiles of MBL Cloud and Drizzle Microphysical Properties Retrieved From Ground-Based Observations and Validated by Aircraft In Situ Measurements Over the Azores, J. Geophys. Res. Atmos., doi:10.1029/2019JD032205, 2020a.

Zheng, Y., Rosenfeld, D. and Li, Z.: Quantifying cloud base updraft speeds of marine stratocumulus from cloud top radiative cooling, Geophys. Res. Lett., doi:10.1002/2016GL071185, 2016.

Zheng, Y., Rosenfeld, D. and Li, Z.: A More General Paradigm for Understanding the Decoupling of Stratocumulus-Topped Boundary Layers: The Importance of Horizontal Temperature Advection, Geophys. Res. Lett., doi:10.1029/2020GL087697, 2020.